PB-216 580

STUDY OF THE PROPERTIES OF AEROSOLS, WITH PARTICULAR REFERENCE TO THE NATURE OF THE AIR-PARTICLE INTERFACE

Alexander Goetz

California Institute of Technology
Pasadena, California

31 August 1958

DISTRIBUTED BY:

NTIS

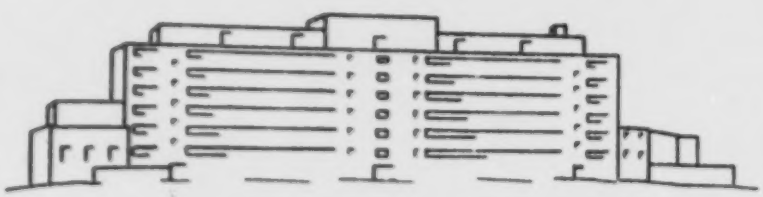
National Technical Information Service
U. S. DEPARTMENT OF COMMERCE
5285 Port Royal Road, Springfield Va. 22151

only one

PB 216 580

The Robert A. Taft
Sanitary Engineering Center
TECHNICAL REPORT
A58-10

LIBRARY
SANITARY ENGINEERING CENTER
CINCINNATI, OHIO



STUDY OF THE PROPERTIES OF AEROSOLS, WITH PARTICULAR REFERENCE TO THE NATURE OF THE AIR-PARTICLE INTERFACE

This technical report is the final report by Dr. Alexander Goetz, California Institute of Technology, under Public Health Service Contract SAPH - 69557.

Reproduced by
NATIONAL TECHNICAL
INFORMATION SERVICE
U.S. Department of Commerce
Springfield VA 22151

U.S. DEPARTMENT OF HEALTH, EDUCATION, AND WELFARE
Public Health Service

1958

96

STUDY OF THE PROPERTIES OF AEROSOLS, WITH PARTICULAR REFERENCE TO THE NATURE OF THE AIR-PARTICLE INTERFACE

(U.S.P.H.S. CONTRACT NO. SAPH-69557)

Prepared for

Air Pollution Engineering Research
Robert A. Taft Sanitary Engineering Center
Cincinnati, Ohio

By

California Institute of Technology
Pasadena, California

Dr. Alexander Goetz

August 31, 1958

Details of illustrations in this
document may be better studied
on microfiche.

U. S. Department of Health, Education, and Welfare
Public Health Service
Bureau of State Services
Division of Sanitary Engineering Services
Robert A. Taft Sanitary Engineering Center
Cincinnati, Ohio

I

F O R E W O R D

The Air Pollution Research and Federal Assistance Act of 1955 (Public Law 159, 84th Congress) authorized the Public Health Service, U. S. Department of Health, Education, and Welfare, to carry on a program of research and technical assistance related to air pollution. As a means of expediting the air pollution research effort under this Program, the Act provides for the letting of research contracts. Contract research, as well as grant research, complements the activities carried on directly by the Public Health Service and by other Federal agencies.

The field of aerosol dynamics is a largely neglected one, in no small measure because of its complexity. One distinguished contributor to the knowledge of airborne particles has been Dr. Alexander Goetz, Department of Physics, California Institute of Technology. In recognition of the importance of particle behavior studies, a contract was initiated with that institution for studies on the application of Dr. Goetz's techniques to problems of air pollution.

The report of the work conducted under this contract, contained herein, is published and made available for the information of individuals and organizations to whom it will be useful.

CALIFORNIA INSTITUTE OF TECHNOLOGY
PASADENA, CALIFORNIA

August 26, 1958

Mr. Frank Tetzlaff
Chief, Air Pollution Engineering Program
Division of Sanitary Engineering Services
Public Health Service
Department of Health, Education, and Welfare
330 C Street S. W.
Washington, D.C.

Dear Mr. Tetzlaff:

Herewith I submit to you in fulfilment of the obligation, under USPHS Research Contract No. SAph-69557, with the California Institute of Technology, Pasadena, Calif., 10 copies of my Final Report, entitled: "Study of the Properties of Aerosols, with Particular Reference to the Nature of the Air-Particle Interface".

Sincerely yours,

Dr. Alexander Goetz
Principal Investigator

AG/lh

III

FINAL REPORT

USPHS RESEARCH CONTRACT SAph-69557

Principal Investigator: Dr. Alexander Goetz

STUDY OF THE PROPERTIES OF AEROSOLS, WITH PARTICULAR REFERENCE TO THE NATURE OF THE AIR-PARTICLE INTERFACE.

Period: 1 January 1958 through 31 August 1958

CALIFORNIA INSTITUTE OF TECHNOLOGY
PASADENA, CALIFORNIA

IV

General Introduction:

The ultimate target of the efforts at this laboratory has been since 1955 to gain more basic understanding of the physical nature of air-borne particulate matter, commonly termed: "aerosols" - particularly of the (statistical) reaction equilibria with trace-components of the suspending atmosphere, of which very little is known.

The significance of such knowledge is seen in the following: If accumulation - ranging from statistical temporary association with the particle surface to adsorbed surface layers - occurs with irritating or toxic gas traces in the atmosphere, an intensification of the physiological effect can be expected, i.e. a synergism between particulate and gaseous components - for combinations suitable for such combination. The physical reason for this intensification is the occurrence of a high topical concentration of irritant molecules upon collision with a physiologically sensitive surface by such "loaded" particles.

Although such synergisms have been reported in the literature, no systematic investigation is known - understandable because methodology and instrumentation in this field are not yet sufficiently developed. -

In the judgment of the author two principal "tools" were missing: an instrument for the separation and size classification of air-borne particles in the range of 5 μ - 0.05 μ (diameter), - the size class predominating in atmospheric aerosols and significant from the public health viewpoint.

Second, a microbiological indicator, which does not involve the statistical fluctuations and the requirements of space and personnel etc. for animal tests or human panels, because these impose serious limitations on the large volume of test data needed.

In 1956 the first successful prototype of an "Aerosol Spectrometer" was built and used subsequently for the size analysis of synthetic and natural "smog".

A significantly improved type was available during the period of this contract. Its performance has been studied - particularly with regard to the possibility of tracing with it associated states between gas components and air-borne particles. This subject forms Part A of this Report.

The "Bacteriological Irritation Analogue" (BIA) test, its instrumentation and application is described in Part B.

Both developments were made early, because without these "tools", the planned systematic studies of the problem of atmospheric synergism were considered unfeasible.

Attached to this Report is an Appendix, containing a theoretical analysis and interpretation of recent animal tests in synergistic aerosols (Amdur - Harvard), because of the close parallelism with the principal subject of this Report.

- - - - - - - -

PART A

THE AEROSOL SPECTROMETER AND ITS APPLICATION

by

Alexander Goetz (Intro.-I.) and Heber J.R. Stevenson (II.-IV)

LIST OF CONTENTS

		Pg.
1.)	Introduction	1
2.)	Basic Principle	2
I.	Design and Performance of the Aerosol Spectrometer	
1.)	Design and Construction	6
2.)	Performance Characteristics	10
3.)	Discussion	13
II.	Particle Size Distribution Determinations	
1.)	Microscope Counts	18
2.)	Surface Reflectivity	19
III.	Applications	
1.)	Controllable Variations of Indicated Particle Size.	21
2.)	Effect on Eye-Irritation from Auto Exhaust	24
3.)	Effect on Plant Damage from Auto Exhaust	27
IV.	Summary	30

- - - - - - - -

1.) Introduction:

In the study of the constitution and the reaction kinetics of systems consisting of suspensions of discrete particles in gaseous fluids, the most serious limitation is the necessity for precipitating the particles in the submicron range (5 μ - 0.05 μ, i.e. the most significant size range for many air pollution problems) out of the air-borne state and separating them according to their diameters (or mass).

This size-classification is particularly important since micro-analytic information about the constituents of aerosols can be seriously misleading because of the cubic relation between particle-mass and -diameter, whereby the presence of a few 1 - 2 μ - particles can obscure the true nature of thousands in the .1 μ - class. For the same mass the latter have an at least 10 times larger surface available for catalytic and/or sorptive interaction with the free molecular components of the suspending gas - but also a 10-fold larger mobility due to the Brownian movement. Hence the same quantity of an irritating substance occuring either as one 1 μ - particle or as 1,000 units of .1 μ diameter becomes in the latter case about 10^4 times more available to a physiologically sensitive surface, such as is involved in eye irritation.

A condition of no lesser significance requires the process of precipitation to least interfere with particle shape, size, and chemical nature. In most separating (e.g. filtration and impingement) methods, the surface of the particle is exposed, after its retention

on the impinger surface or within the filter matrix, to the shearing action of the passing gas stream. It can be assumed that this shear disturbs the dynamic equilibrium of loosely sorbed or condensed components on the particle surface (probably most effective in irritation) to a considerable extent.

A method which preserves the original physical and chemical status of the formerly suspended particle is thus particularly important for the subsequent evaluation of the true nature of an aerosol.

- - - - - - - -

2.) Basic Principle:

The approach to a practical solution of the problem is based on the application of centrifugal acceleration to a continuous flow of the gaseous suspension under test. It appears logical to base the principle of discrimination between particle and surrounding gas on the sole, universal physical difference between both, i.e. the specific gravity; furthermore, the use of centrifugal force, suitably applied, should least alter the nature of the precipitated particle from that of the air-borne state.

In principle this force can be applied by streaming the gas through a helical or spiral channel long enough for all particles to reach its outer wall which forms the locus of the particle deposit. The size classification would thus be caused by the difference between the radial velocity components of the large and small particles. The former will all be deposited near to the

channel entrance, the latter further toward the end.

The practical realization of this principle meets with serious mutually conflicting requirements.

a.) Because of the extremely small individual masses involved (10^{-12} - 10^{-16} gr) the centrifugal acceleration, i.e., the flow rate through the helix, has to be very large indeed in order to produce a radial velocity component sufficient for the particle to reach the outer wall of the helix within the time of its presence in the channel. This leaves the alternative between very large flow-rates or excessive channel lengths (amounting to several miles for .2 μ particles).

b.) The radial trajectory of each particle (across the gas flow in the channel) in the centrifugal field must be subject to Stokes' law in order to be predictable. This requires <u>laminar</u> flow in the channel (i.e. low Reynold's number) which bars the application of high flow velocities.

c.) The laminar flow condition necessitates the existence of a boundary layer at the channel walls, virtually resting thereon. Hence a particle, after entering this layer, will no longer be subject to centrifugal force and it will lose its velocity in radial direction, if its momentum (i.e. mass) is small. Consequently very small particles may never reach the wall, because they cannot penetrate the boundary layer.

This dilemma can be overcome by moving the wall <u>with</u> the fluid, i.e. by rotating the helical channel around its longitudinal axis in the direction of the flow. If the helical channel rotates at

high angular velocities, the flow within is exposed to a large centrifugal field, even for low flow velocities in the channel.

Consequently the flow can be kept within the range where the law of Stokes-Cunningham controls the velocity of the particles' radial movement, in spite of the application of large centrifugal fields and short channels; also the principal difficulty, presented by the resting boundary layer, disappears since the centrifugal field is acting in it as well as upon the particle deposit on the wall. The latter presents a significant advantage by itself in effecting a fixation of the particles without the use of adhesive agents (which by the act of adhesion can alter the nature of the particle surface). If these conditions are fulfilled in sufficient approximation, the locus of particles deposited on the outer channel wall should permit a determination of their size distribution, since the 3 factors determining the centrifugal force acting upon the particles of various masses can be measured and controlled, namely the geometry and angular speed of the helix and the relative velocity between flow and channel.

The basic element of such an instrument is thus a rotor of circular cross-section normal to its axis of rotation, the periphery of which carries a helical groove covered by a removable flexible surface, the latter sealing the groove to an air-tight channel during operation and being available after removal from the rotor for subsequent investigation of the deposit as a flat surface.

A considerable advantage of a rotating helix channel is its "self-propelling action" upon the fluid filling the channel. The momentum imparted by the latter upon the former causes the gas to

flow through the channel (impeller action) in a direction determined by the pitch and sense of rotation of the helix. The requirement for an auxiliary pump for moving the gas through the channel can thus be avoided, and the flow velocity can be controlled accurately by partial blocking.

The performance of such a device is obviously determined by the geometry of the rotor: If it is a <u>cylinder</u> the centrifugal acceleration is constant over the entire length of the helical channel. This causes an unfavorable "dispersion" of the particle sizes, for deposit of the smallest fraction will occupy a much wider range on the periphery of the helix than the larger fraction - due to the inverse quadratic relation (d^{-2}) in Stokes' law.(1) If the rotor represents a <u>cone</u> and the channel thus a conical helix of constant pitch and cross-section, the centrifugal force acting on each particle increases with the increasing radius of the cone. Hence the small fraction will be subjected to a larger force than the large particles in case the flow enters the channel on the tip and leaves at the bottom of the cone. Depending on the angle of the cone, the "dispersion" of sizes in the deposit can be controlled and be converted from the quadratic ($\sim d^{-2}$) to a closely linear ($\sim d^{-1}$) and thus more favorable dependence on the particle diameter.

- - - - - - - -

A.I. DESIGN AND PERFORMANCE OF THE AEROSOL SPECTROMETER

1.) Design and Construction:

The conical rotor type has been employed in two models of spectrometers, one of which (Model D) was employed mainly for large volume collection of aerosols or their removal (fig. 1), because of its relatively large volume capacity (20 - 30 liter/min). The other type (Model E) is a commercially available instrument(+), which follows the basic design of (Model D), but has a lesser sampling

(+) Instrument Development & Mfg. Co., Div. of Peterman Enterprises, Inc., 3018 East Foothill Boulevard, Pasadena 8, California.

capacity (4 - 8 liter/min), however specially suited for analysis of aerosols.

Since the experimental work outlined in the following was done predominantly with the latter, the design and performance of the (Model E) is discussed in the following:

The Aerosol Spectrometer consists of 2 units. One unit represents the centrifuge, the other is a chassis with the controls, connected with the instrument by a 5-core cable.

Fig. 2a shows a vertical cross-section, fig. 2b photographic views of the centrifuge unit.

The rotor (R) is a cone, which carries on its outer surface 2 1/2 turns of a double helix, forming 3 of 4 walls of 2 independent helical channels. These channels are sealed by an exactly fitting

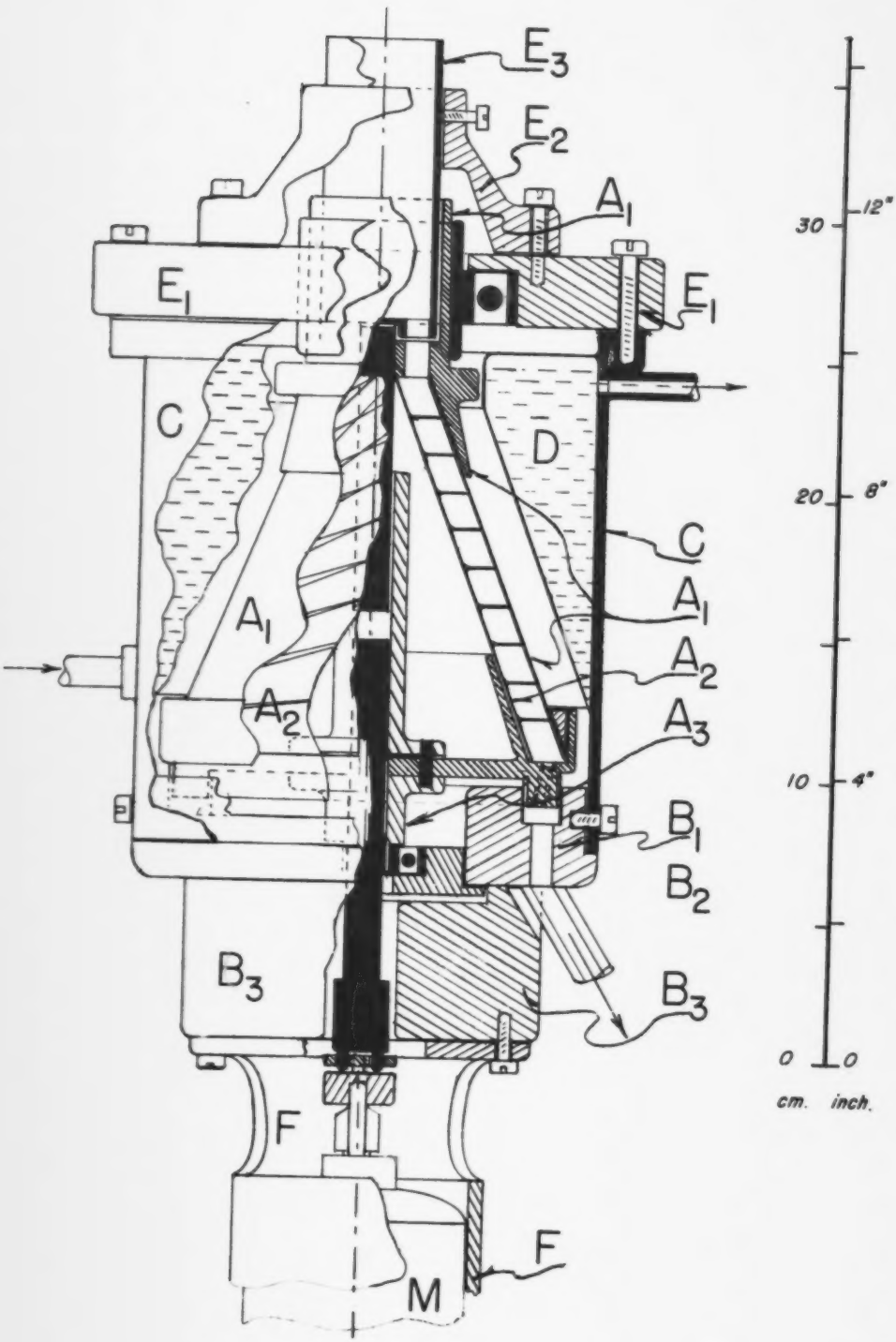


Fig. 1 Vertical cross-section of Aerosol Spectrometer Model "D".

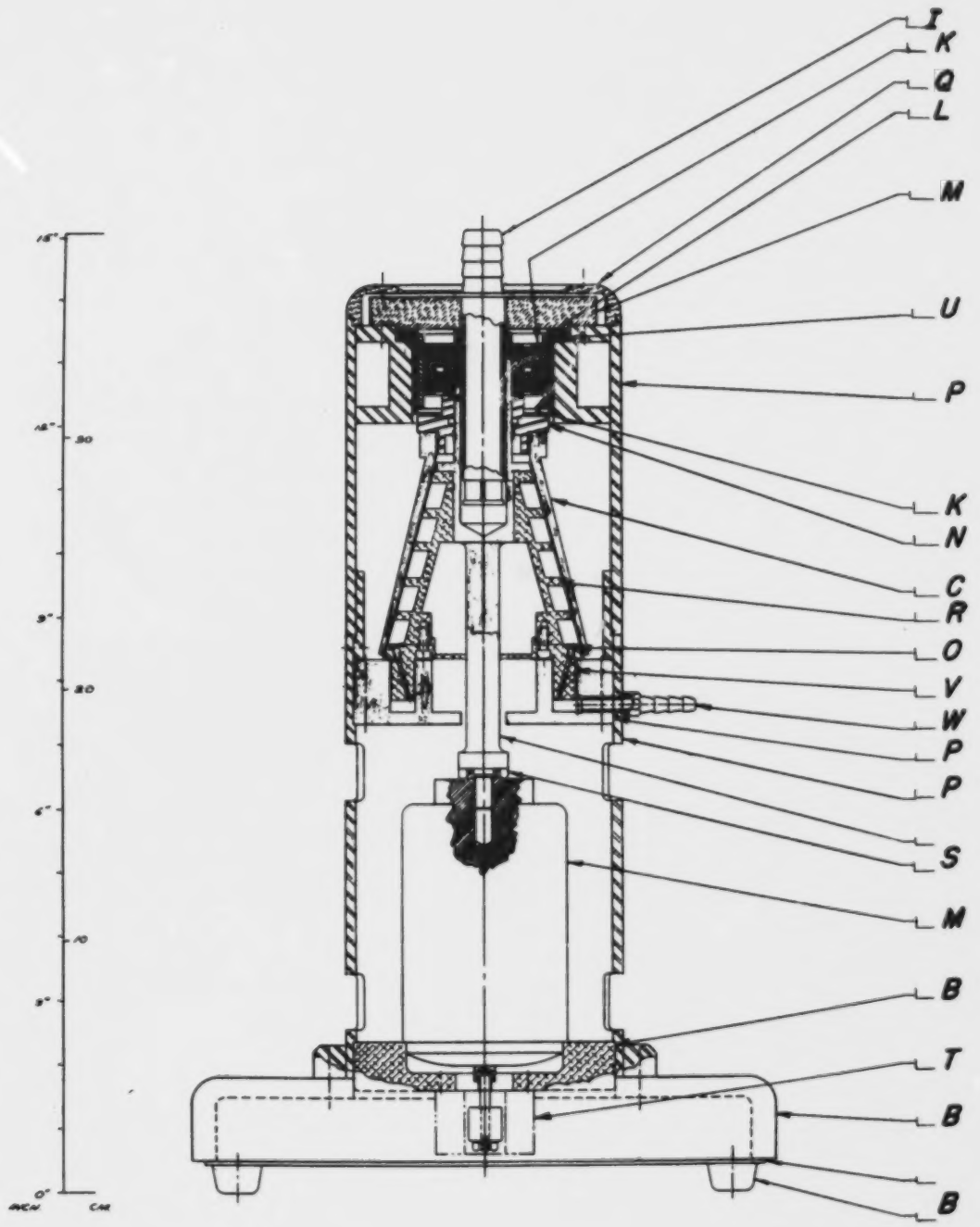


Fig. 2a Vertical cross-section of Aerosol Spectrometer Model "E".

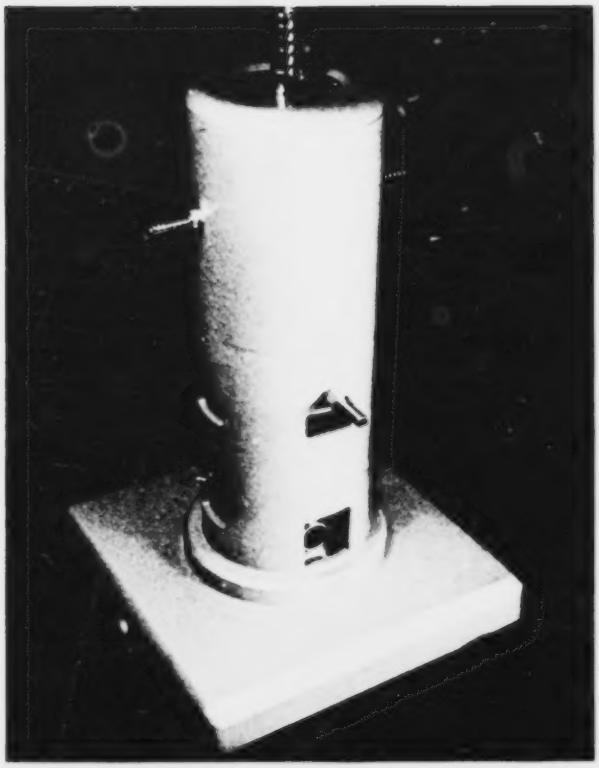


Fig. 2b Photograph of Aerosol Spectrometer Model "E".

conical cup (C), the interior of which is lined with a removable foil, whereupon the particles are deposited. (C) is pressed against (R) by the nut (N).

The rotor is driven by the electric motor (M), mounted in the base (B). (M) drives the armature (T) of an electric tachometer. (R) is supported and driven by a massive drive shaft (S).

The aerosol enters through the (resting) intake tube (I), which fits loosely into the hollow center at the upper end of the cone. One port at the upper end of each channel is exactly opposite to the openings at the lower end of (I). In this manner the air flow is guided with least disturbance into the rotor.

(R) is guided at the upper end by the air lubricated "frictionless" bearing assembly (consisting of the constituent parts [I, K, L, M, U]).

The entire assembly is contained in a heavy removable shell (P), which carries on top a lucite window (Q) for stroboscopic observation of the top bearing, i.e. for the independent check of the speed of rotation.

After having passed the channels, the air flow leaves through exchangable orifices (O) which seal each channel and restrict the flow velocity. The orifices lead the gas through the ring shape base (V) of the rotor into a close fitting annular recess in the base, which communicates with the outlet tube (W). This design renders the air after the channel passage available to analysis etc. with least chance of subsequent contamination by the aerosols of the motor etc.

A conical jacket for water cooling (not shown) fills the inner space in the shell within a small distance from the rotor. It absorbs the heat generated by the air-friction on the rotor shell and permits the adjustment of the temperature of the peripheral channel surface, where the particles are deposited.

The chassis contains a variable auto-transformer unit for the speed regulation, volt- and ammeter (for checking the operating conditions of the motor), the tachometer, switch and fuses (in the primary and the secondary circuit); it is connected by cable and plug with the power supply.

The characteristic dimensions of the instrument are the following:

Motor: Brush-type, AC or DC 135 V. max., 1/4 HP, max. speed 26,000 rpm = 433 rps.

Rotor:

height: ..... 2.63" (= 6.68 cm)

full angle of cone (α): ..... 30°

pitch: ..... 1.085" (= 2.76 cm) per turn

turns (n): ..... 2.5 (= 5 π)

Channels:

number of helical channels (ν) 2

horizontal diameters of:

inner wall at inlet ports (D_1) .... 1.03" (= 2.6 cm)

outer " " " " (D_1') .... 1.53" (= 3.9 cm)

inner " " outlet (D_2) .... 2.50" (= 6.34 cm)

outer " " " (D_2') .... 3.00" (= 7.62 cm)

width of thread (w): ..... .128" (= .325 cm)

" " channel (vertical) (b): .. .416" (= 1.06 cm)

depth " " (horizontal) (c):.. .250" (= .635 cm)

cross-section of channel

$[A = b \cdot c \cdot \cos(\alpha/2)]$: ..... .10 sq. inch (= .650 cm^2)

overall channel length from

in- to outlet (L_o): ..... 15.9" (= 40.4 cm)

volume of each channel ($V_c = A \cdot L_o$) .. 1.59 cu. inch. (= 26.3 cm^3)

limiting orifices ("jets")

(one pair each) with bores: ..... 0.02" (= .05 cm)

0.04" (= .10 cm)

0.06" (= .20 cm)

From these data those constants can be derived, which determine the performance of the instrument, namely:

the <u>flow velocity relative to the channel</u> as:

$$v_r(\text{cm/sec}) = \frac{F \cdot 10^3}{60 \cdot \nu \cdot A} = 12.9 \cdot F \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (1)$$

(F = air flow i. liter/min)

the <u>Reynold's Number:</u>

$$R^x = \frac{\rho}{2\eta} \cdot v_r(b+c) = 5.66\, v_r = 73 \cdot F \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (2)$$

the <u>time of residence</u> in the channel:

$$\tau_o(\text{sec}) = \frac{\nu \cdot V_c \cdot 60}{F \cdot 10^3} = \frac{3.15}{F} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (3)$$

The time of residence of a particle in the channel prior its deposition at the distance (L) from the channel entry:

$$\tau_L(\text{sec}) = \tau_o \cdot L/L_o = 2.48 \cdot 10^{-2} \cdot \tau_o \cdot L \quad \ldots\ldots\ldots\ldots\ldots\ldots \quad (3a)$$

The centrifugal acceleration [$\gamma \cdot g$, i.e. as multiple of the gravity constant (g)] has for each value of (N) a minimal ($\sim D_1$) and a maximal ($\sim D_2'$) value:

$$\gamma = \frac{2 \cdot \pi^2}{9.8} \cdot 10^{-2} \cdot D \cdot N^2 \text{ (for D, i. cm; N, i. rps)} \quad \ldots\ldots\ldots \quad (4)$$

or:

$$\left.\begin{aligned} \gamma_{min} &= 2 \cdot 10^{-2} \cdot D_1 \cdot N^2 = 5.2 \cdot 10^{-2} \cdot N^2 \\ \gamma_{max} &= 2 \cdot 10^{-2} \cdot D_2' \cdot N^2 = 1.52 \cdot 10^{-1} \cdot N^2 \end{aligned}\right\} \quad \ldots\ldots\ldots\ldots \quad (4a)$$

$$\frac{\gamma_{max}}{\gamma_{min}} = 2.94$$

- - - - - - - -

2.) Performance characteristics:

The strict mathematical analysis of the particle trajectories within the channel, which determine the place of deposition on the outer wall as function of (N), the speed of rotation, (d) the Stokes' diameter of the particle, and (v_r) is quite involved and outside the scope of this report.(1)

The distribution of the deposit along the channel wall for particles of the diameter (d) is continuous until a distance from the channel entry is reached (L_d) where <u>all</u> particles are deposited. Obviously (L_d) <u>in</u>creases with <u>de</u>creasing (d), and $\left[L_d(d)\right]$ <u>de</u>creases with <u>in</u>creasing (N). Furthermore $\left[L_d(d)\right]$ depends on (τ_o), i.e. on (v_r) which is controlled by the orifices. -

Empirical calibration with polystyrene latex aerosols of exactly defined shape and diameter - available in the range ($1.2\ \mu \geq d \geq .08\ \mu$) - results for each value of (N) and (v_r) in $d(L_d)_{N,\ v_r}$.

It is convenient to use only few values of (N) and (v_r) for operation selected for optimal dispersion within certain size ranges.

For 3 values of (N) and 3 sizes of orifices, 4 principal $d(L_d)$ - functions have been used which cover the range ($3\ \mu \geq d \geq .05\ \mu$).

Table I. lists the values for (N), (O = orifice aperture) and thus resulting values of P, τ_o, γ_{min}, γ_{max}, and the maximal and minimal diameters (d_{max}, d_{min}) capable of complete elimination and resolution.

TABLE I.

Unit	N rpm	O .001"	F lit/min	τ_o sec	γ_{min} g	γ_{max} g	d_{max} μ	d_{min} μ	Symbol
	6,000	60	2.1	1.5	520	1,520	3	.75	A
	12,000	"	6.5	.48	2,800	6,100	2.5	.45	B
	"	40	4.5	.70	"	"	1.5	.25	C
	24,000	20	7.5	.42	8,300	24,500	1.0	.04	D

The maximal value of γ obtainable with this instrument approaches 30,000 at N = 26,000.

Fig. 3 represents the corresponding functions in log-coordinates, each line resulting from a calibration with at least 2(A), at most 5 different monodisperse glass- [for 5 μ] and latex aerosols in the range from (5 μ ≥ d ≥ 0.08 μ) while the range (0.08 μ ≥ d ≥ 0.04 μ) is extrapolated. -

Fig. 4 shows typical deposit patterns of such aerosols on (black) paper foils after removal from the rotor cup.

On a plane surface the helical channel walls, i.e. the pattern of the deposit, form 2 parallel Archimedian spirals; while the locus of all points of equal distance (L) from the channel entry are circles concentric with the circular boundary of the foil. (L) is therefore defined by the length of the spiral path from the entry to the intersection with a circle forming the boundary of the deposit.

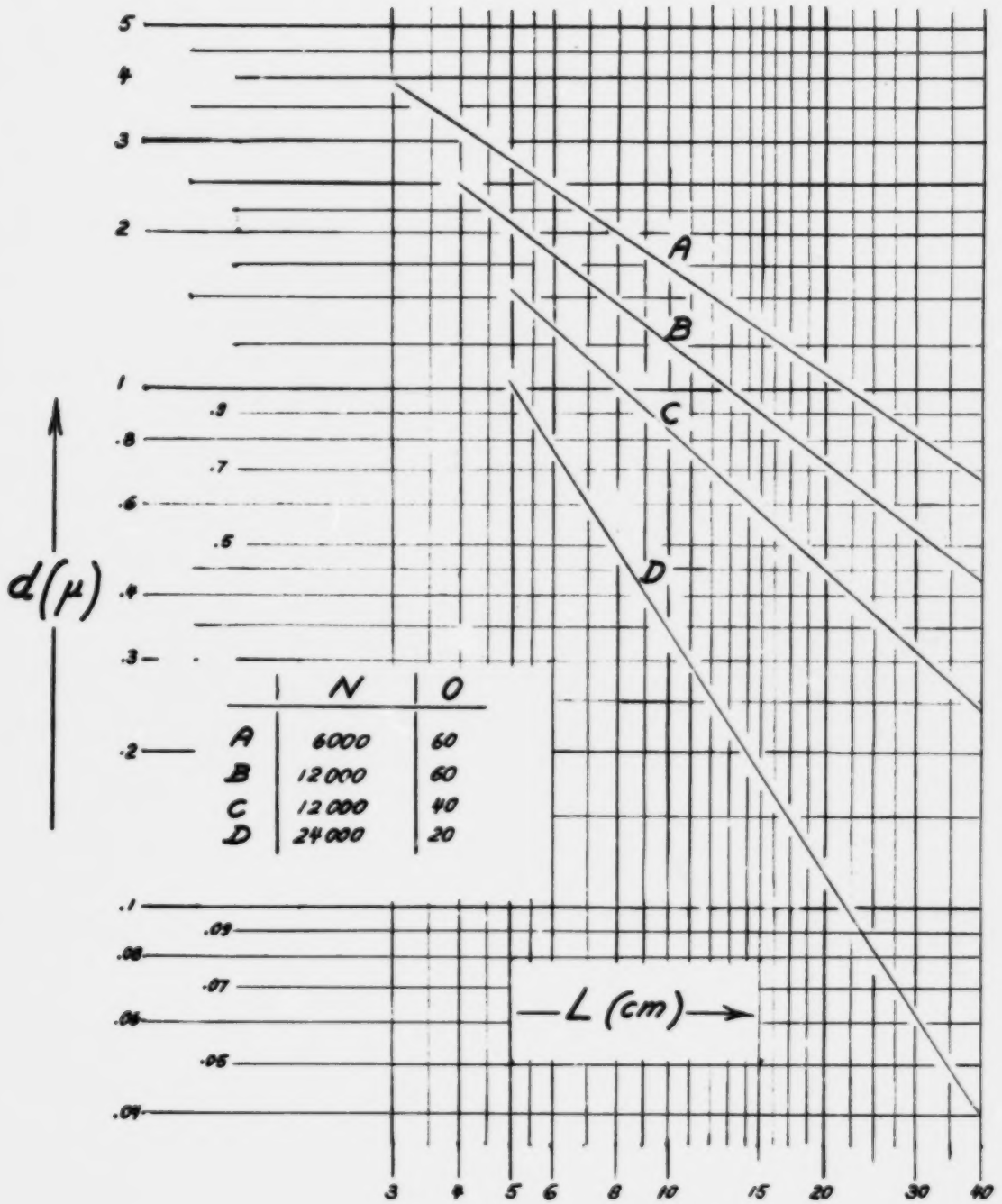


Fig. 3 Calibration curves showing particle size as a function of distance along the channel for several speeds and air flows.

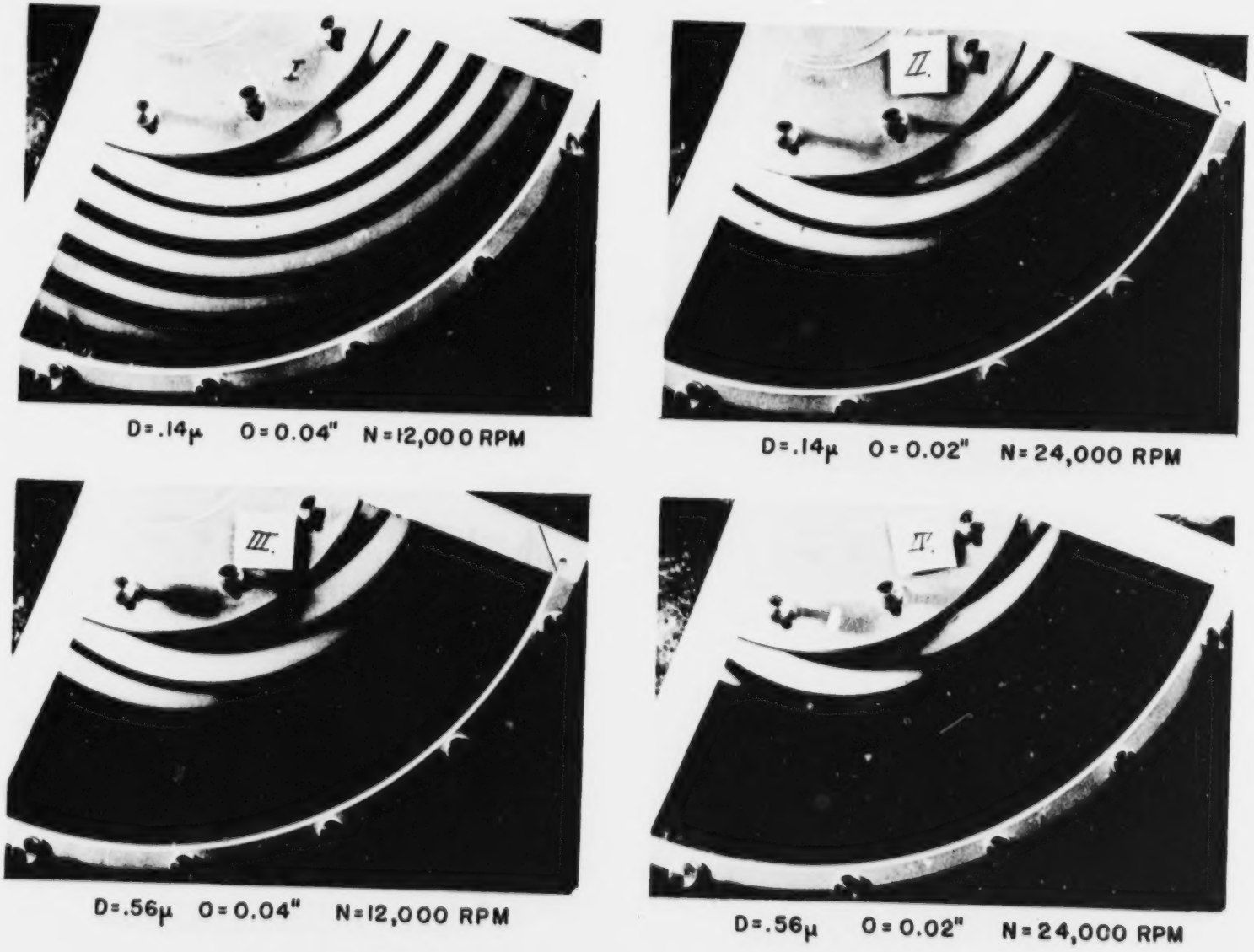


Fig. 4 Typical deposit patterns of polystyrene latex on black paper.

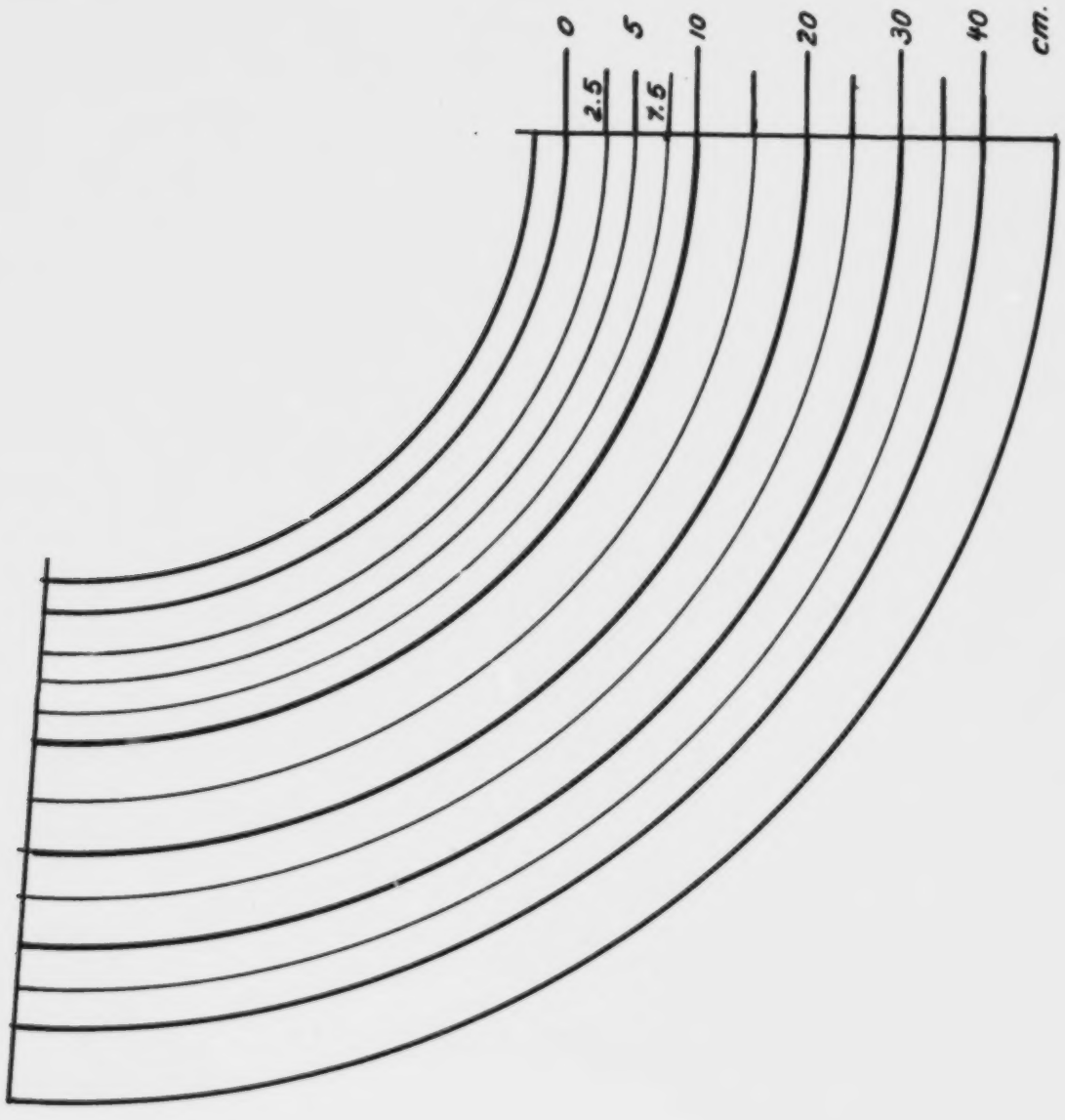


Fig. 5 Calibration diagram giving length of particle deposit along channel.

The determination of (L) is facilitated with a "spiral scale" (fig. 5) - photographically copied on transparent film, in order to permit superposition over the foil.

It presents a family of concentric circlesections the radii of which vary according to the length (L) of the spiral at the point of its intersection with the circles. They are indicated at intervals of L = 2 cm up to 40 cm. In conjunction with the calibration diagram (fig. 3), the length (L_d) establishes the maximal Stokes' diameter (d) of the particles deposited at this channel length, under the particular operating conditions (N and O).

The analysis of the particle-size distribution in polydisperse aerosol requires the knowledge of the density distribution of a particular particle size (d) along (L) for ($L \leq L_d$) (2). The original procedure was based on microscopic counts and has been replaced to some extent by measurements of the variation of surface reflectivity. (see II 1, 2)

- - - - - - - - - -

3.) Discussion:

It is a particular aspect of the Aerosol Spectrometer principle (i.e. of the centrifugal separation of particulate matter from a laminar air flow) that it renders the locus of deposition of a particle with known dimensions a source of information about its condition prior to the time of its deposition. In other words: if the diameter (d) in a monodisperse aerosol is known and (by calibration) the

corresponding value of (L_d), any deviation from this value, i.e. ($L_d' \gtrless L_d$) under the same mechanical operating conditions, with respect to (N) and (O), must be indicative of a change of the "Stokes' diameter" of the particles from (δ) to (δ').

This offers an opportunity, unavailable heretofore, of investigating the physico- chemical conditions in the gaseous environment of the air-borne particle, which are conducive of such changes of (δ). -

Prior to a closer discussion of the physical interpretation of (δ) a condition should be mentioned because of its significance for this particular aspect:

The motion of the particle across the channel/relative to (v_p) the air flow along the latter is but a small fraction ($\sim 10^{-2}$) of (v_r), i.e. of the order of 1 cm/sec. Hence the "air-shear" the particles - especially the smallest particles - are exposed to is minute, thus an interference with gas molecules loosely associated with the particle surface should be quite small.

Under conditions favorable for such accumulations the "Stokes' diameter" will be altered and must be traceable by a change of (L_d). -

Stokes' law (in its simplest form) predicts for spheres that ($v_p \sim \rho \cdot d^2$), where (ρ) the density, and (d) the (geometrical) diameter of the sphere(+). Consequently:

(+) For polystyrene particles (ρ = 1.05$\sim$1.0), hence (δ = d) under normal conditions.

$$v_p \sim \delta^2 = \rho \cdot d^2 = \frac{m}{k \cdot d} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (1)$$

if (m) is the mass of the particle and (k) a geometrical "shape factor" (for a sphere: $k = \pi/6 = .52$). -

Assumed: an aerosol with a partial pressure of a gas (e.g. H_2O) sufficiently large for causing condensation on the particle, this condensation to result in a "hull" of liquid of the density (ρ_c), [in other words the particle acts as condensation nucleus, its density: ($\rho_n = \rho_c$)]. In this simple example the particle will be deposited sooner, hence:

$$L_d' < L_d;\ \delta' = d' = \delta + \Delta = d + \Delta \quad \ldots\ldots\ldots\ldots\ldots\ldots \quad (2)$$

where ($\Delta/2$) represents the thickness of the condensate around the air-borne particle.(+)

(+) It is noteworthy that (L_d') is independent of the fate of the condensate after the deposition of the particle, e.g. its subsequent disappearance by evaporation, absorption etc.; i.e. (L_d') allows to reconstruct the condition of the particle in the air-borne state. -

---

Less obvious is the effect on (δ) in case a loose association, e.g. of polar trace components, with the particle would occur(3): If such a "hull" moves <u>with</u> its nucleus, but would - because of its gaseous constitution - not increase (m) significantly, the mass of the complex would be ($m' = m$), while ($\rho_n \gg \rho_c$). For this case eq. (1) results in:

$$\left[\frac{\delta'}{\delta}\right]^2 = \frac{k \cdot m' \cdot d}{k \cdot m \cdot d'} = \frac{d}{d'} = \frac{d}{d+\Delta} \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots \quad (3)$$

and (since for $\rho = 1$, $\delta = d$)

$$\delta' = \left[\frac{d^3}{d+\Delta}\right]^{1/2} \quad \ldots\ldots \text{ and } \Delta = d\left[\left(\frac{d}{\delta'}\right)^2 - 1\right] \ldots\ldots\ldots\ldots \quad (4)$$

Eq. (4) indicates: $(d = \delta > \delta')$, i.e. a gaseous hull causes a <u>decrease</u> of the Stokes' diameter; a <u>larger</u> L_d-value should be observed than for the not associated particle [despite the larger, hydrodynamically effective, diameter $(d' > d)$] . -

This effect should be more marked for small than for large particles, if under the same conditions equal values of (Δ) may be assumed (+), for two reasons:

(+) Such an assumption may be not realistic in view of the increase of volatility (vapor pressure) on the radius of (convex) curvature of the particle surface (Kelvin's law). For a more detailed discussion of this point, reference is made to (3).

---

a.) Eq. (4) indicates $\delta/\delta' = \left[1 + \Delta/d\right]^{1/2}$. For example: if $\Delta/2 = 3 \cdot 10^{-6}$ cm, i.e. 1/2 - 1/3 of a mean free path of a gas molecule, δ' would result in: .47 μ (for $d = .5\ \mu$ - i.e. a 6% reduction), while for $(d = .12\ \mu)$, $\delta' = .098\ \mu$ - i.e. the same Δ would cause a 20% reduction.

b.) The relative velocity of the particle (v_p) is <u>less</u> for <u>small</u> then for <u>large</u> (d) under the <u>same</u> operating conditions of

the instrument. For very loose associations (s.a.) the air shear will thus be less for small than for large particles resulting in larger (Δ) for the smaller particles.

This effect can be compensated by altering the operating conditions so that both particle sizes experience the same (v_p), i.e. have the same (L_d).

Furthermore, it can be predicted that the temperature should have a marked effect on (Δ), for the state of such associations is likely to be thermally quite metastabile.

- - - - - - - -

The above discussion appeared justified - despite its speculative elements - because the effect of ($\delta' \lessdot d$) has actually been observed and reproduced for synthetic latex aerosols by particular variations of conditions - as described in a later chapter (A,III,1).

- - - - - - - -

A.II. PARTICLE SIZE DISTRIBUTION DETERMINATIONS

1.) Microscope Counts:

The microscopic method has proven successful for counting particles even below 0.1 μ by using dark field illumination from above, but as nearly parallel to the surface as possible. The particles have been counted when collected either on highly polished chrome-plated brass shim stock or on glossy black paper. The limiting factor is the structure of the imperfections of the surface. The point is reached where one has to judge which are collected particles and which are imperfections. Fig. 6 shows several sizes of uniform polystyrene latex spheres deposited on a chromium surface as seen through the microscope. The lines of the counting reticule are also seen.

The apparent size of the particles is of no importance in the determination of particle size in this procedure. The smaller sizes, being so much less than the resolving power of light microscopy, have no apparent difference in size, only a difference in light intensity which would be very difficult to evaluate. However, since size is determined by position, particles are counted only in terms of their position and the size distribution determined analytically. In the work described in New York and San Francisco,[2] the counting method was used for analysis and the procedure is described there.

- - - - - - - -

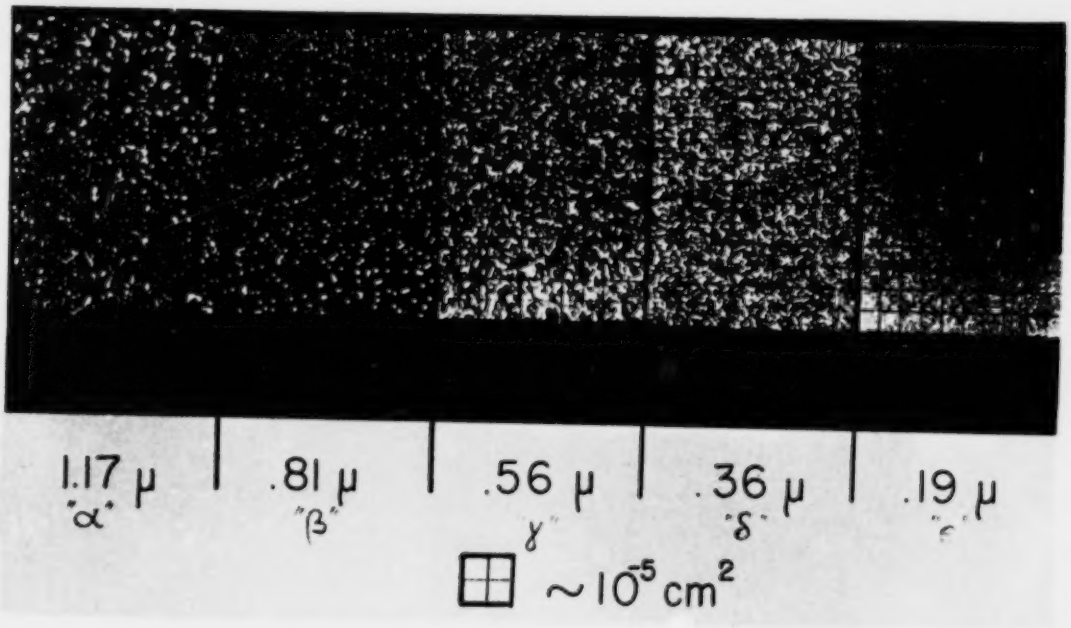


Fig. 6 Photomicrographs in dark-field reflected illumination of calibrated polystyrene aerosol deposits.

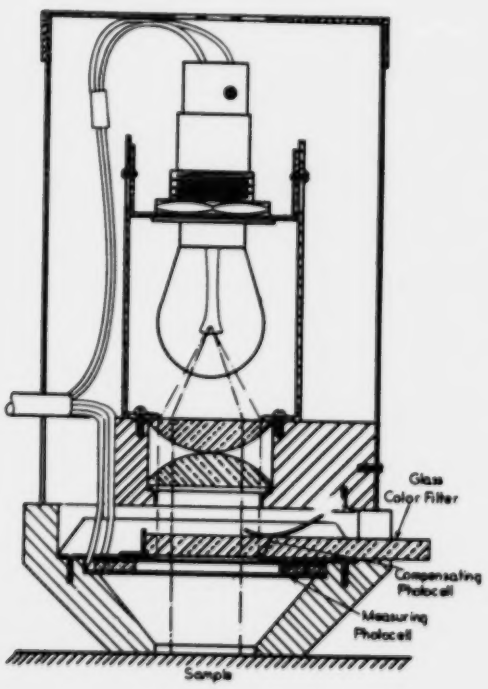


Fig. 7 Search unit for photoelectric reflectivity meter.

2.) Surface Reflectivity:

In order to decrease the time required for analysis and to avoid tedious microscope counting, a procedure has been developed using diffuse surface reflectivity. Fig. 7 shows the search unit which when used together with a galvanometer gives reflectivity readings. The problem in this procedure was positioning of the sensing unit laterally as well as longitudinally, since the concentration varies in both directions. In order to accomplish this, a mask with marks each centimeter was made from card board to lay over the latex deposits so that the search unit could be placed reproducibly on the deposit without smudging it and to determine the position along the channel. A spiral strip was cut from the mask to correspond to the channel to be measured so the latex deposit could be seen through the mask. A second mask the same shape as the face of the sensing head was constructed with a rectangular hole 3 by 10 millimeters cut in black paper. By moving this second mask from position to position on the first mask which is stationary over the deposit, accurately reproducible areas may be measured. The sensing head is placed in the same position in the second mask each time and the amount of light scattering is read on the galvanometer scale with 100 divisions. The readings are generally reproducible to one division. The scattered light is sensed by an annular barrier layer cell which surrounds a beam of light from an incandescent bulb to the surface being measured. A second barrier layer cell turned upward is included in the circuit to provide an adjustable zero. The annular cell does not receive light from an optically smooth surface since this would be specularly reflected

back along the beam and through the center of the annulus. The diffusely reflected light, on the other hand, does arrive at the ring, and is measured by the galvanometer.

The problems with reflectivity are somewhat different than with microscope counting in that the increased light from larger particles does enter into the readings. Also the quantity of sample required with the technique used so far is considerably greater than that required for microscope counting. Fig. 8 shows a comparison of counting vs. percent reflectivity. In the range which it is possible to compare, the relationship is very linear, extrapolates to zero, and the ratio of reflectivity is very nearly the first power of the ratio of the diameters. This may be due to the fact that the sensing cell receives a ring of light from each particle which is proportional to the first power of the diameter. The ratio of the sizes illustrated is 2.1 whereas the ratio of reflectivity is 1.9. Fig. 9 shows the result of plotting reflectivity vs. channel position for various sizes and types.

Some of the disadvantages of the present method may be overcome with a procedure that is being developed here using a device which will automatically follow the Archimedian spiral produced by the channels, and by using a photoelectric cell in the microscope. This will make more precise determination of the profiles possible both longitudinally and laterally. It will have the disadvantage, however, of making imperfections of the supporting surface more important. The moderately high concentrations and relatively large area observed (30 mm^2) smooth out variations - perhaps desirable as well as undesirable.

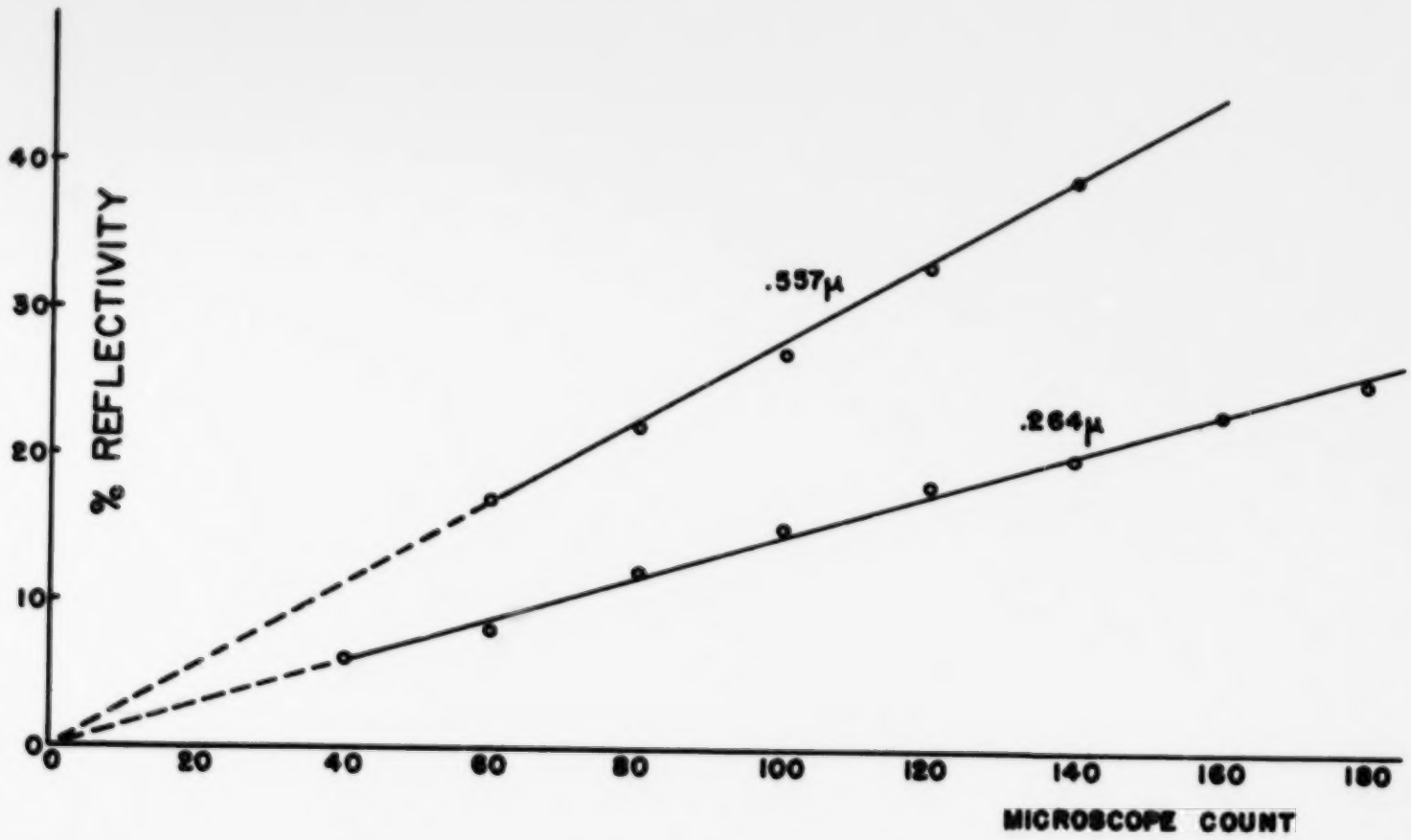


Fig. 8 Reflectivity as a function of count for two particle sizes.

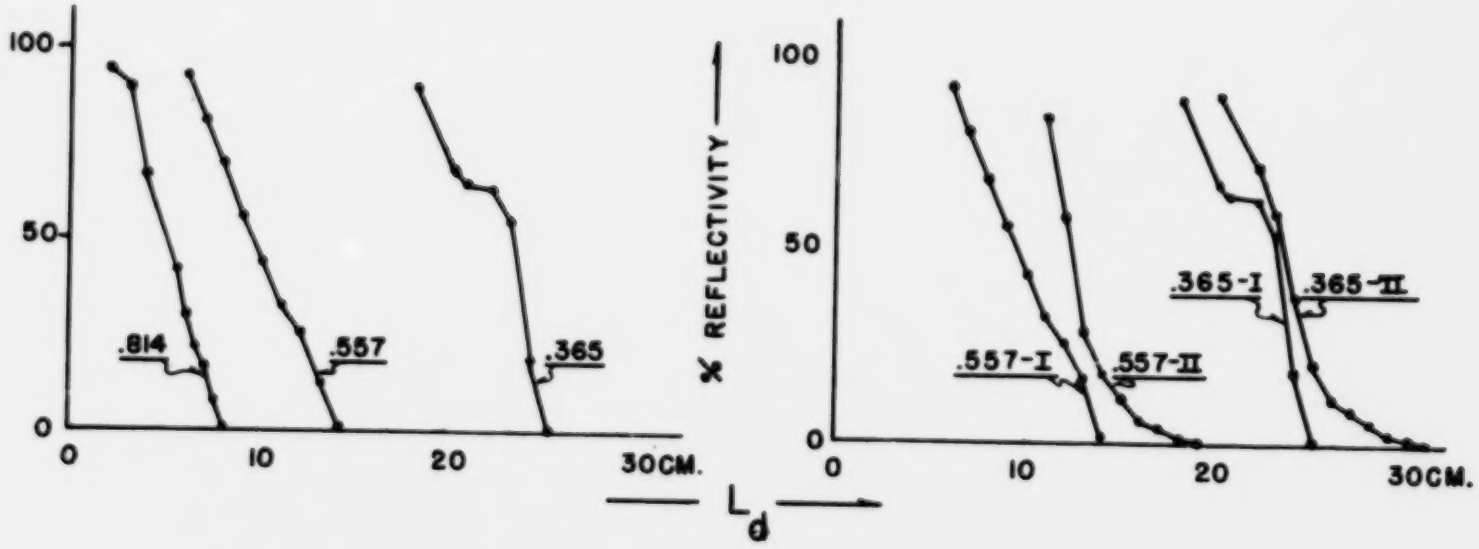
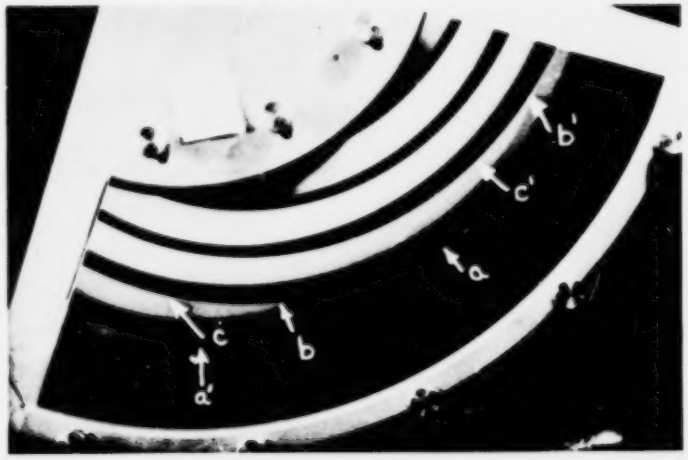
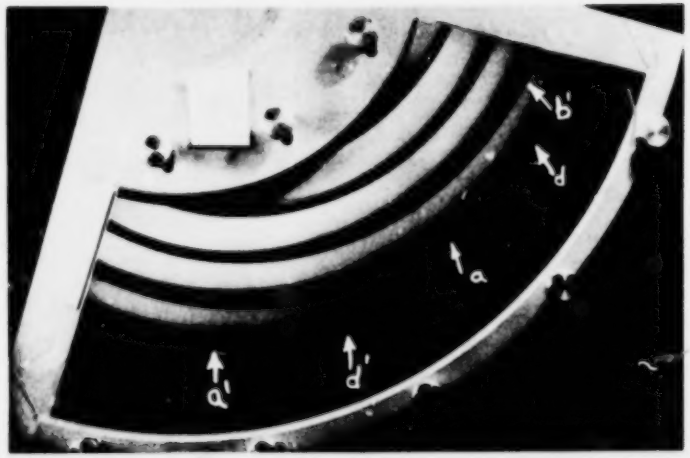


Fig. 9 Plot of reflectivity vs. channel position for several sizes and types.





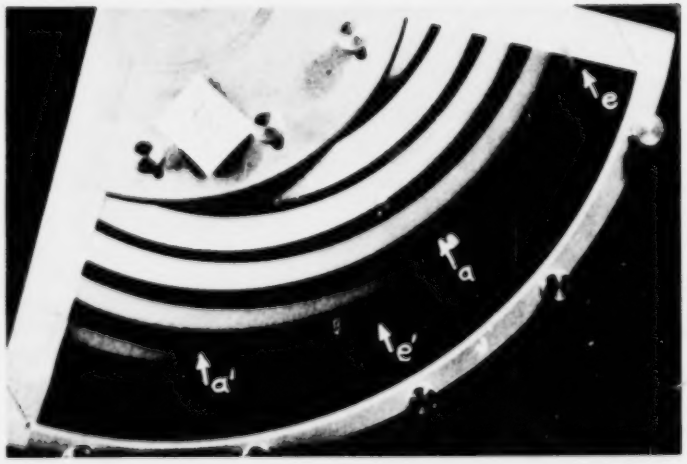


Fig. 10 Types of deposit patterns - primes indicate second channel; a - terminus of type I (shown on II and III for reference); b, c - subboundaries; d, e extended termini.

For the studies observing the apparent change in size of latex particles or any studies where an adequate quantity of sample is available, the reflectivity method is clearly the method of choice. For studies of particles collected from polluted air or from irradiated auto exhaust, more investigation must be performed to determine whether microscope count can be replaced.

- - - - - - - - -

A.III. APPLICATIONS

1.) Controllable Variations of Indicated Particle Size:

The length of deposit of a particular particle size can be increased although the same speed of rotation and air flow are maintained by changing some of the other conditions. As may be seen in fig. 9, this is not simply a shift to a longer deposit, but a change in the character of the deposit. For the purposes of this report we have called the two general profiles produced types I and II. Fig. 10 shows the 0.365 μ particles aerosolized from n-propyl alcohol as a type I. Aerosolized from water is a type II. In photo III it was sprayed from methanol and shows an even greater effect. The main characteristic of the type II profile is the fact that it is an extension to the type I profile. Type I has a sharply defined terminal boundary with up to 3 subboundaries (ghosts) preceding the final boundary. Type II, on the other hand, shows evidences of a corresponding final boundary, but, depending upon the degree of the effect, this boundary is obscured by an additional deposit (tail) to another, less

definite terminus further along the channel. Another characteristic of type I is the fact that the deposit narrows gradually after the first few centimeters of deposit. The type II deposit also shows the initial narrowing, but the additional deposit which characterizes type II usually fills the channel.

The conditions under which a type II profile can be produced are to some extent controllable. A typical type I profile is produced when the latex particles .365 μ in diameter are aerosolized from water into a small (7 liter) chamber before being led into the centrifuge. If the aerosol is led directly to the centrifuge through a T connection to vent any excess and prevent a change in the pressure at the inlet of the centrifuge, the profile shows a tendency toward a type II. If the latex is aerosolized from methyl alcohol, through a T, to a warmed-up centrifuge, a typical type II is formed. If the box is used in the place of the T, or the centrifuge is not warmed up, the profile is intermediate between I and II.

The difference just described also holds with larger particles .557 μ in diameter. In one test on a different instrument still larger particles .814 μ appeared not to show a difference when sprayed from water or methanol, but this should be repeated on the first instrument. Smaller particles, on the other hand, showed the type II profile when sprayed from water through a T, but showed the type I when sprayed from a HCHO solution into the small chamber.

When the speed of the centrifuge was increased from 12 to 18 thousand RPM, thefirst particles described (d = .365 μ) showed a type

II profile whether sprayed from water or methanol.

The production of the type II profiles with the .365 μ latex from methanol was first studied using a centrifuge equipped with a water jacket (110 W) which could be used for cooling. The jacket was not filled with water for these experiments and the increased air resistance seemed to make the instrument warmer than instruments without the water jacket. When a second instrument (109) was used for making the deposits, with everything else being the same, spraying from methanol gave type I instead of type II profiles. The difference apparently was the lower temperature in the instrument without the jacket.

A possible explanation which seems to fit the various facts depends upon a concept of decreasing the Stokes' diameter ($d\sqrt{\rho}$) at the same time as increasing the actual diameter (d) by surrounding the particle with a layer of gaseous material (water vapor, methanol vapor, HCHO, etc.) which maintains a density (ρ) more nearly that of gas than liquid or solid. For example, if a 1 micron particle of $\rho = 1$ has a gaseous hull giving a total diameter of 2 microns, the combined particle would have only 1/8 times the density since the mass would remain approximately the same with gas densities being about 0.001 g/cm^3. Hence, the Stokes' diameter would be $\sqrt{1/8} \cdot 2 = .7$ μ or less than the solid central part or nucleus of the combined particle. This means that although the particle has increased in size it will come out later or farther along the aerosol spectrometer channel and hence could explain the appearance of the type II profile.

If this is the explanation, then conditions which are conducive to the formation of a gaseous hull would produce the type II

profile. The most important factor in a less than saturated atmosphere would be the length of time from the formation of the aerosol particle in the nebulizer till its deposit in the aerosol spectrometer. When more time is permitted, as in aerosolizing into the chamber, the hull has more chance to disperse. The temperature of the spectrometer would have the opposing effect and cause the hulls to persist by increasing the vapor pressure. The methanol, having a relatively high vapor pressure causes the hull to last longer. Normal propanol with less vapor pressure than methanol produces even less effect than water.

It should be pointed out that although these hulls maintain very low density, they no longer are Maxwell-Boltzman type gases and the term "gasoid"-hulls might be more appropriate.

The effect of coriolis type forces in a conical rotor appears interesting and remains to be evaluated.

- - - - - - - -

2.) Effect on Eye-Irritation from Auto Exhaust:

The following table, taken from previous work and partially quoted in the paper in the APCA Proceedings (2) is given here to illustrate the effective reduction of eye irritation from irradiated auto exhaust under proper conditions, namely cooled centrifuge, and fresh collection surface.

Date	Irritation Chamber	Irritation "D"	Ratio	Reaction time hours	Collection Surface
11.12.	13.6	5.6	.41	2	fresh saran
11.14.	10.4	13.6	1.31	4 1/4	loaded saran
12. 5.	9.8	7.1	.73	4 3/4	painted surface
12.10.	23.2	9.6	.41	2	fresh saran
12.17.	19.2	7.0	.36	2	fresh saran

These tests were done at the Stanford Research Institute with their car irradiation chamber and irritation panel.(4) The irritation index is an arbitrary scale with a theoretical maximum of 24. Earlier than 11/12 measurements of eye irritation of exhaust passed through the centrifuge showed no reduction. The centrifuge was at that time not cooled.

The 11/14 test was left out of our previous papers because of the different experimental conditions, but is probably highly significant.

On this day, a very long collection had been made in order to have a large sample for chemical analysis. The collection surface was therefore heavily loaded. It has also been observed that during the early part of an experiment, the irritation is greater than it is later. Hence, the exhaust arriving at the centrifuge earlier undoubtedly had a higher index. In any case the surface was now

saturated and in spite of the cooled surface, the irritant reevaporated and gave an increased index. On 12/5 the sample for analysis was removed before the panel was exposed, on 12/10 and 12/17 fresh saran surfaces were put on the centrifuge just before the test and the reduction ratio was very closely that of 11/12 when fresh saran was used.

In March two more tests were conducted under conditions that produced less aerosol.

Date	Panel	Calculated Residence Time	Irritation Chamber	Irritation "D"	Ratio
3.17	1	3.4 h	11.4	6.8	.6
	2	1.9	12.0	14.0	1.2
	3	1.1	17.3	18.6	1.1
3.19.	1	3.4	12	12	1.0
	2	1.9	12	9.3	.8
	3	1.1	22.6	20	.9

These tests indicate that eye irritation can be produced without producing much aerosol in which case the centrifugal reduction is small.

By combining all the foregoing data, it seems reasonable to conclude that particulate matter is not essential to eye irritation. However, under some conditions, a large eye irritation index is associated with production of a large amount of aerosol. Under these conditions, the irritant is apparently condensed on the particles

since centrifugation can reduce the irritation. The irritant is easily re-evaporated if the collection surface is either warm or saturated, and under conditions, not yet explained, where irritation is produced without production of aerosols, centrifugation is not effective in removing irritation.

This probably explains the results of Neal Richardson and Wilbur C. Middleton(5) that fiber filters are not effective in removing irritation. The irritants could be present as condensed droplets, but although momentarily collected on the filter they would re-evaporate possibly recondensing downwind of the filter. The filter would enhance re-evaporation since the temperature would be slightly higher than ambient and the pressure to the lee would be at least slightly reduced.

- - - - - - - -

3.) Effect on Plant Damage from Auto Exhaust:

In order to determine the extent to which aerosols play a part in the damaging of plants by irradiated auto exhaust as produced in the LAAPCD greenhouse, several experiments were performed in which the irradiated exhaust was passed first through the Model "D" Aerosol Spectrometer to remove particles larger than 0.05 μ. The plant damage which resulted was compared with the damage resulting from exposure to the gases coming directly from the irradiation chamber.

The source of exhaust is a Fairbanks-Morse 2-cylinder motor-

generator with a 660-watt load. A portion of the exhaust is mixed with a flow of filtered air of 0.6 m^3/min. into a chamber of 11 m^3 thus making the calculated turnover time about 18 minutes.

Two plant exposure boxes were constructed of masonite with a removable glass top. Their volume is about 35 liters each. Into these may be set a rack containing up to 9 pots containing young plants to be exposed. These may be connected in series with the aerosol spectrometer in any position. For the first three experiments, the spectrometer was between the boxes. Following the final box was a smaller box containing a thermometer and a hygrometer.

The chamber is irradiated by sunlight and is not cooled, hence in bright sun the temperature often reaches 120° F during the course of an experiment. For this reason, a cooling jacket was constructed through which ice water was circulated surrounding the chamber air as it flowed to the first plant box.

The Model "D" was also cooled with ice water so that the air in the second box was approximately the same temperature as in the first box. On the first day the boxes were in the sun and were about 96° F. For the next few experiments the boxes were under an umbrella and the temperatures were in the 70's.

Recorders for oxidant, CO, NO and NO_2 were in operation during the experiments and grab samples were taken for hydrocarbon, ethylene, acetylene and infrared analyses.

For an experiment with the two plant boxes in series without the aerosol spectrometer between, in order to determine to what extent the plants themselves remove the phytotoxic factor, a blower of the

same capacity was used following the boxes to avoid using the spectrometer as a blower only.

For the final experiment, the hydrocarbon content was raised as high as possible to get plant damage rapidly, so that several experiments could be done in one day. This time the plant boxes were kept cool by draping them with wet cloths rather than shading them.

Samples of the irradiated auto exhaust aerosols both direct and through the Model "D" were collected on the Model "C" aerosol spectrometer to determine the particle size distribution and the effectiveness of the removal by the Model "D".

The results were quite consistant. In all of the experiments at least some of the plants showed typical damage. In no case was there less damage after removal of the aerosols by the aerosol spectrometer. In most cases there was even a slight increase in damage but probably not enough to be of significance. The collected aerosols appeared to be homogeneous particles about .1 micron in size. The Model "D" exhaust was better than 95% particle free.

From this it may be concluded that the phytotoxic agent is - at least under the conditions of this experiment - gaseous. Some caution must be expressed, however, since the turnover time in the chamber is less than 20 minutes. Would the phytotoxic agent condense if it had more time? An experiment to test this was unsuccessful. The fact that no plant damage has been produced with the S.R.I. chamber using longer residence times may indicate instability of the phytotoxic agent.

- - - - - - - -

A.IV. SUMMARY FOR PART A.

The aerosol spectrometer has proven to be an effective tool for the study of the role of particulate matter in air pollution. It has demonstrated that both eye irritation and plant damage can be produced by gases alone. It has further shown that when eye irritation is associated with high values of aerosol - which may be the usual case - the irritation can be reduced by the removal of particles if they are not permitted to re-evaporate. It has shown also that the association of the irritant with the aerosol is undoubtedly in the condensed state since an increase in Stokes' diameter is produced. This is in contradistinction to the decrease in Stokes' diameter concurrent with an increase in actual diameter of particles of known size with a "gasoid"-hull, i.e. a layer of molecules of very low density but moving along with the particle. This phenomenon appears to be producible under various conditions.

- - - - - - - -

Subtexts to Fig. 1 - 10 (Part A)

Fig. 1 Vertical cross-section of Aerosol Spectrometer Model "D"

Fig. 2a Vertical cross-section of Aerosol Spectrometer Model "E"

Fig. 2b Photograph of Aerosol Spectrometer Model "E"

Fig. 3 Calibration curves showing particle size as a function of distance along the channel for several speeds and air flows.

Fig. 4 Typical deposit patterns of polystyrene latex on black paper.

Fig. 5 Calibration diagram giving length of particle deposit along channel.

Fig. 6 Photomicrographs in dark-field reflected illumination of calibrated polystyrene aerosol deposits.

Fig. 7 Search unit for photoelectric reflectivity meter.

Fig. 8 Reflectivity as a function of count for two particle sizes.

Fig. 9 Plot of reflectivity vs. channel position for several sizes and types.

Fig. 10 Types of deposit patterns.

BIBLIOGRAPHY FOR PART A

1.) A. Goetz. An Instrument for the Quantitative Separation and Size-Classification of Air-Borne Particulate Matter Down to 0.2 Micron. Rev. Geofisica Pura E Applicata, Proc. II. Intern. Symp. Condensation Nuclei, Basel-Locarno, 1956, 36, 49-69, 1957.

2.) A. Goetz and H.J.R. Stevenson. The Aerosol Spectrometer Its Theory, Construction and Application to the Analysis of Exhaust and Atmospheric Aerosols. APCA Proceedings, San Francisco Nov. 1957.

3.) A. Goetz. Zur Physik der Luftkolloide im Submikronen-Bereich. Heidelberger Physiker Tagung, October 1957. (In Press)

4.) E. Schuck. Eye Irritation from Irradiated Auto Exhaust. Report No. 18, Air Pollution Foundation Southern Calif. Lab. Stanford Research Institute, 1957.

5.) N. A. Richardson and W. C. Middleton. Evaluation of Filters for Removing Irritants from Polluted Air. Univ. of Calif., Los Angeles, Report 57-43, June 1957.

PART B

THE BACTERIOLOGICAL IRRITATION ANALOGUE (BIA) TEST

by

Alexander Goetz in partial collaboration with Noel Tsuneishi

LIST OF CONTENTS

		Pg.
1.)	General Purpose	1
2.)	Basic Operating Principle	2
I.	The Rotary Impactor	
1.)	Purpose	5
2.)	Design	5
3.)	Modes of Application	8
II.	Generation of Test Aerosols	10
III.	Quantitative Relations	12
IV.	Bacteriological Procedure	
1.)	The Working Principle of the Concentrometer	16
2.)	Preparation of the Bacterial I-Strip	20
V.	Results of (BIA) Tests	
1.)	Automobile Exhaust	22
2.)	Tobacco Smoke	23
3.)	Synthetic Aerosols	24
	Conclusion	28
	Acknowledgement	
	Bibliography	

- - - - - -

B. THE BACTERIOLOGICAL IRRITATION ANALOGUE (BIA) TEST

1.) General Purpose:

The development of a test procedure, which provides for an easy, reproducible analogue to physiological irritation on a microbiological basis is considered of principal significance, although a general parallelism between, e.g. bacterial growth inhibition and physiological reactions in a human, may not be anticipated. However, it appears reasonable to assume that for specific systems, e.g. atmospheric oxydants etc., analogous reactions could be produced, provided suitable procedures could be developed for this type of irritant.

Another promising application of such a method would be the quantitative study of synergistic phenomena between gaseous and particulate atmospheric components, i.e. between a gaseous irritant in extreme dilution and a (per se neutral) aerosol. This type of study depends to a large extent on biological indicators, requires, however, also a very large number of experiments, each involving as well numerous biological individual "units", to minimize statistical fluctuations. -

The use of microorganisms (bacteria, yeast cells, etc.) seemed thus promising, inspite of their evolutionary distance from the mammalian forms because of:

a.) their unlimited availability,

b.) ease of standardization,

c.) wide tolerance to conditioning by temperature, nutrient composition, etc.

d.) absence of statistical fluctuation due to large numbers,

e.) availability of membrane culture technique, which allows the isolation of the cells from its nutrient environment during the test exposure,

f.) rapid development of the irritation "signal" (8-24 hrs.)

g.) low test cost and space requirements compared with animals or human panels.

In view of these promising, basic features, a method was designed 1957, and suggested to the USPHS, Robert A. Taft Sanitary Engineering Center, Cincinnati, which was subsequently evaluated by the authors and independently by Mr. H. F. Clark, Sen. Bacteriologist of the above institution, in a purely exploratory manner, Since it proved workable and indicative of an analogy with eye-irritation (auto exhaust, smog), additional efforts were spent during the period of the contract on improvement of technique and instrumentation as well as on exploratory applications to synthetic synergistic aerosols. -

- - - - - - - -

2.) Basic Operating Principle:

The specific reason for this particular procedure is to effect a direct contact between the aerosol particles and the cell surface of the bacteria - i.e. with minimal interference by a liquid layer shielding the former physically or chemically (buffering

action) - contrary to the aerosol contact with a liquid bacterial suspension.

This contact is established by impacting the aerosol onto bacterial cell layers of known density. To obtain reproducible results, volume, flow rate, impacting velocity, and time of exposure must be accurately controlled. Such layers of cells can easily be produced on the surface of membrane filters by previous passage of a diluted 24 hrs culture of organisms. After the aerosol exposure the membrane is cultured according to standard procedures.(1)

Principally, the irritant intensity (J) could be evaluated by two alternative procedures: By exposing a membrane surface:

a.) carrying a uniform bacterial density (N = const.) to a topically varying volume of the impacting aerosol, by then determining the position of a zone on the membrane, where growth inhibition first occurs, and correlating this zone with the aerosol volume which had contacted it.

This method was considered impracticable because of the virtual impossibility to accurately reproduce (N) in different tests.

b.) carrying a bacterial density (N), which varies along one direction (x), to a constant flow of aerosol; (J) is then defined by the reduction of growth development along (x).

The latter alternative was selected because the concentrometer method(2) facilitates conveniently the automatic production of membrane deposits with logarithmically varying density $\left[\lg N = A + \frac{x}{k}\right]$. This form of distribution eliminates within a wide margin the uncertainty due to the variation of (N) (c.f.).

- - - - - - - -

Obviously the BIA test requires the development of several component procedures. Since the concentrometer and associated bacteriological techniques had been previously developed by the author and his collaborators, they were available and required but minor modifications. A major effort during the contract period however, was required for the construction and performance evaluation of the impacting device - as well as of a procedure to generate quantitatively defined aerosols for the BIA test. -

Each such development is described in detail in the following.

- - - - - - - -

B. I. THE ROTARY IMPACTOR

1.) Purpose:

In principle, the instrument is a surface impactor of the slit-type, its impacted surface is represented by the periphery of a rotating cylinder, the width of which is about twice the length of the slit. The purpose of this construction is:

a.) to minimize the extension of the contact of the air flow through the slit beyond the area represented by the slit aperture on the impacted surface. This is accomplished by channeling the air-path so that only a small fraction of the whole strip, directly opposite to the slit, is contacted at any one time.

b.) to obtain on the surface, so impacted, a continuous sequence (i.e. record) of exposure to the aerosol. This is effected by rotating the cylinder at constant (or varying) speed, while a constant air-flow is maintained through the slit.

2.) Design: (Fig's. 1 and 2)

The instrument is housed in the shell (A), the open (left) side of which is closed vacuum-tight (O-ring) by the plate (B), removal of which provides access to the rotor (D). The inlet tube (C_1) ends in (0.03 cm x 0.63 cm) slit, formed by the shape of the lower end of (C_1) and by the tight fitting plate (C_2), held to (C_1) by screws. This arrangement permits access for adjustment and cleaning - and for the reproducibility of the slit-width upon reassembly. The distance of (C_1) is adjustable relative to the periphery of (D) by the vertical movement of (C_1) in tubular extension of (A). The latter forms a wide, slot-like opening for the passage of the air-flow after impaction to the outlet (N),

which is connected to the vacuum pump, manometer, flowgauge etc. - Moreover, the penetration of (C) through (P) causes only one (left) side of (C_1) to communicate with (N). (see arrows).

The shaft (E_1), carrying the rotor (D), is supported by the bearing (F), which forms a vacuum-tight (O-ring) gland. Since the assembly is thus a vacuum-tight unit, suction at (N) will result in an air intake only at (C).

The shaft (E_1) carries (D) with a sliding fit, its relative position to (E_1) is fixed by a pin (E_2), driven by (E_1), hence (D) is easily withdrawn or inserted after removal of (B) without affecting the (critical) distance between rotor-surface and slit.

(E_1) is driven by exchangable synchronous motors (M) of 4 or 12rph, so that the exposure of the cylindrical section of (D) to the air flow from the slit is 12 min. and 4 min.respectively.

To define the relative position of rotor and slit at the start, a friction clutch transfers the motion of (M) through the plate (G_1) to (E_3) and thus to (E_1). The (slight) pressure required between (G_1) and (E_3) is provided by 2 bronze spring plates (K) which also form the support of (M) at the base plate. (E_3) carries the handle (G_2), the turning of which (against the friction of the clutch) adjusts the position of (D) - at the same time (G_2) acts as indicator for the position of (D) on a circular scale at (H). -

The rotor surface forms a cylinder over an angle of 255° of its circumference, the remainder is relieved to provide room for the clamping device of the test strip (I-strip), which covers

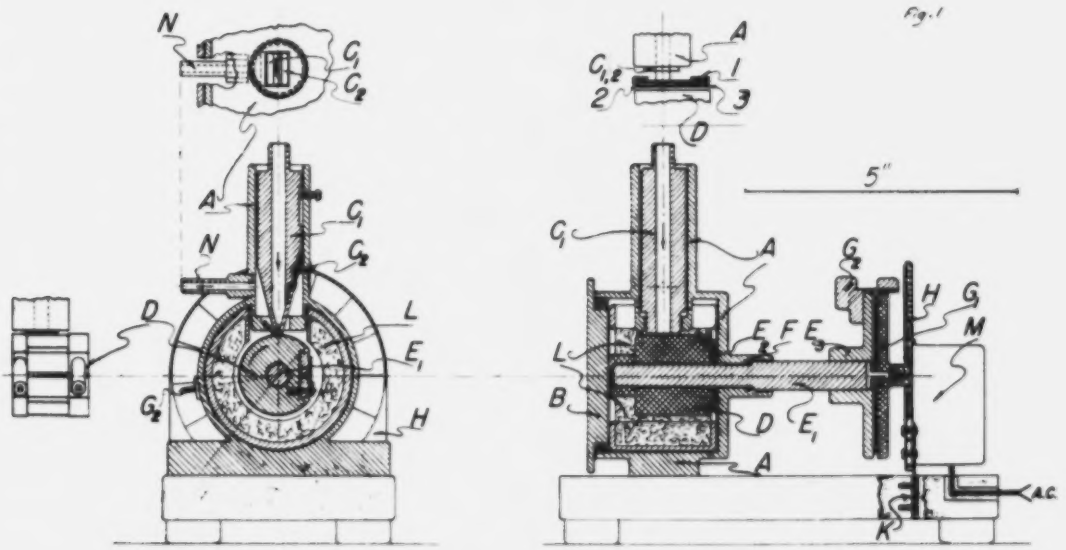


Fig. 1 Vertical cross-sections and partial cuts of Rotary Impactor.

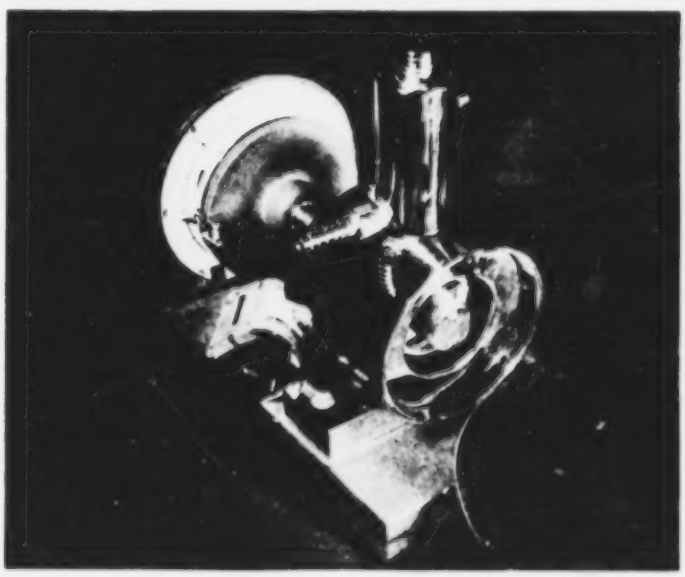


Fig. 2 Photograph of Rotary Impactor.

the cylindrical section. This clamping device is designed to hold the I-strip under longitudinal tension (to assure its constant distance from the slit) - also to render touching the I-strip surface unnecessary, while applying it onto, or removing it from the rotor, - important, when chemical reagents or biological preparations (bacteria) are carried on the I-strip. An auxiliary device (fig. 3) which supports clamp and I-strip, while detached from the rotor, facilitates this operation. -

The width of (D) is about twice as wide as the slit-length, in order to provide for control purposes an equivalent, non-exposed area on the I-strip.

For applications, where the non-exposed parts of the I-strip should be protected against drying, a moisture donor (L) is provided, consisting of a removable cylindrical shell, exposing a thick, well-fitted lining of hydrophilic felt toward (D), which is saturated with water, before each test. -

The slit - when acting as limiting orifice (Δ_p = .5 at) has maximal flow capacity (F_i = 10 liter/min) for air [at T ~ 26°C].

At a rotor speed of 4 (12) rph, the maximal total test sample amounts to ca. 120 (40) liters [4.2 (1.4) cu.ft.] of air. The total area on the I-strip exposed to impaction is (A_c = 5.2 cm^2), the air volume, impacting per unit area of the I-strip during one revolution, [i.e. the time (t)] is thus: ($F_i \cdot t/A_c$) and has accordingly the maximal concentration factors of 23 (7.7) liters/cm^2; or 5.2 (1.7) cu.ft./sq.inch.

The slit aperture is (.63 cm x 0.03 cm = $2 \cdot 10^{-2}$ cm^2),

hence at (F_i = 7 liter/min), the generally used flow-rate, the flow velocity, prior to impaction, amounts to: 56 m/sec = Mach .17.

- - - - - - - -

3.) Modes of Application:

The impactor has been designed for the analysis of aerosols by three basically different modes of application, depending on whether (D) is rotated at constant, or at varying angular velocity.

1.) At constant velocity, the deposit on the I-strip (e.g. when carrying a reagent for specific aerosol components) will present the variation of the general or specific aerosol density within the duration of the test. Consequently the rate of dispersion of an aerosol from an instantaneous source can be determined. (Striking of match, puff of smoke, are simple ways of demonstrating the principle. c.f.)

2.) If the I-strip is prepared so that the surface concentration of the reagent varies along the former according to a predetermined function, (preferably a log-function) the method can serve for the quantitative indication of specific (not varying) aerosol components, by direct comparison with a standard strip derived from an aerosol of known concentration. The preparation of such strips with a logarithmically varying deposit is facilitated with the concentrometer.

This method has been applied for test strips carrying bacterial cells, upon which irritating aerosols were impacted. (c.f.)

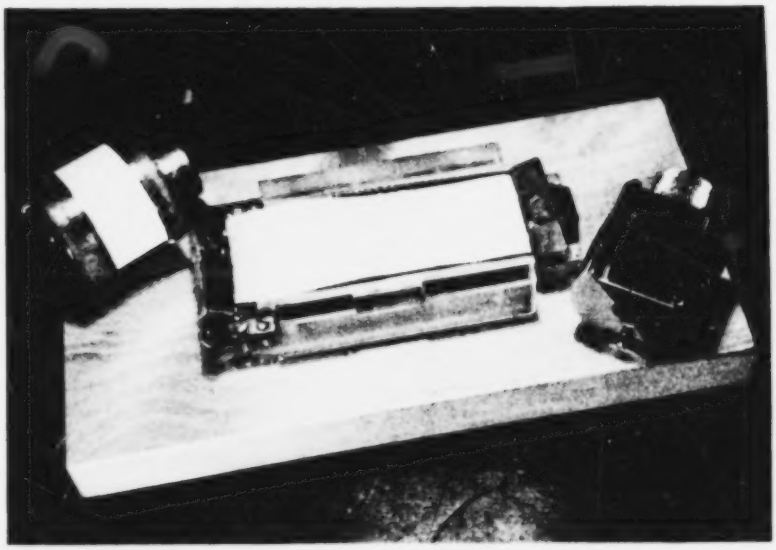


Fig. 3 Lucite holder for mounting I-strip from monodisperse latex aerosols (d = .26 μ).

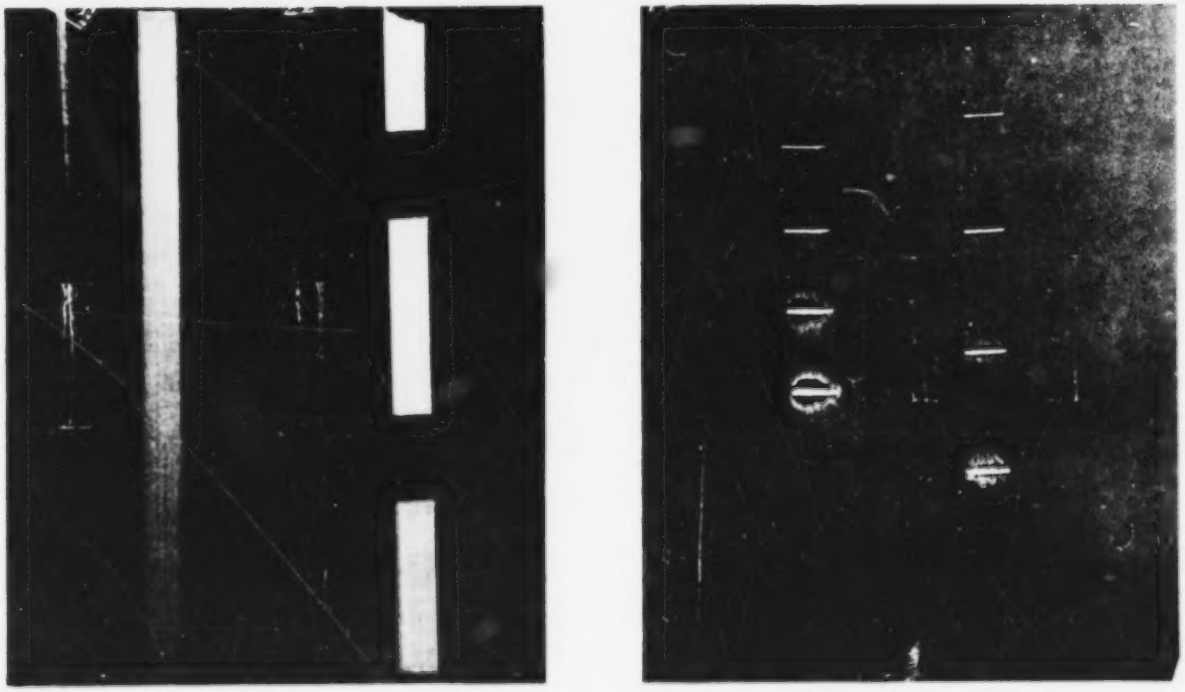


Fig. 4 Rotary Impactor I-strip with impactor patterns: a.), for continuous (left) and intervalled (right) aerosol in flow (note sharp definition); b.), impaction patterns on stationary rotor (right) and analogous pattern on conventional slide impactor.

3.) If the angular velocity of (D) during impaction varies (e.g. from fast to slow speed), particularly, if the variation with time is logarithmic, a result similar to 2.) is obtained on I-strips which carry the indicator at uniform surface concentration.

- - - - - - - -

Impaction Pattern

As demonstration (fig. 4a) of the performance of the rotary impactor, a monodisperse latex aerosol (d = .36 μ) was generated (c.f.) and impacted on the surface of a glossy black paper, corresponding to γ_i = 10 and 20; in another test the aerosol was cut off for 1 min. during one revolution in order to show the sharpness of the intersection.

The difference in pattern of the deposit between this and that of the usual construction, which involves impaction on a plane surface is shown by analogous deposits in the stationary system where the exposure time varied as: (1 : 3 : 10 : 30). These patterns (fig. 4b) demonstrate a closer restriction to the slit projection, also the absence of a "halo"-like secondary pattern characteristic for the usual (plane) surface impinger. -

This deposit pattern is significant in view of the inhibition pattern on bacterial membrane cultures, caused by the impaction of irritant aerosols.

- - - - - - - -

B.II. Generation of Test Aerosols

For the exploratory investigation of synergistic effects of toxic irritant gas components, the following method was employed (fig. 5):

The components of the aerosol were combined in an aqueous solution, resp. suspension and dispersed by a commercial venturi-type nebulizer, activated by compressed air of controlled pressure (Δp) at the inlet of the nebulizer.

The resulting air flow (F_n, i. liter/min) was determined by rotameter, the dispersed liquid (M_n, i. gr/min) by weighing the nebulizer before and after a defined time interval of operation. This determines the reproducible "nebulizer-constant" $\left[W = M_n/F_n, \text{ i. gr/liter}\right]$ for aqueous dilutions or suspension of subsequent aerosol components, provided that the time of operation is short enough to avoid significant depletion of the liquid. (W) was found to be of the order of $\left[10^{-2} \text{ cm}^3/\text{liter}\right]$ for $\left[V_n = 9 \text{ liter/min at } \Delta p = 12 \text{ cm Hg}\right]$, hence at the longest time of operation (12 min) ca. 1 cm^3, i.e. only 20% of the liquid were nebulized. The variation of (W) during this interval remained within 5%. A fresh filling from stock solution for each operation of the nebulizer was considered mandatory - as well as a thorough cleaning.

The exhaust of the nebulizer (N) was led either directly to the rotary impactor (s.a.) or after passage through a container (C), depending on the residence time required between generation and impaction of the aerosol. The volume of this container provided for an "age" of ca. 45 sec. -

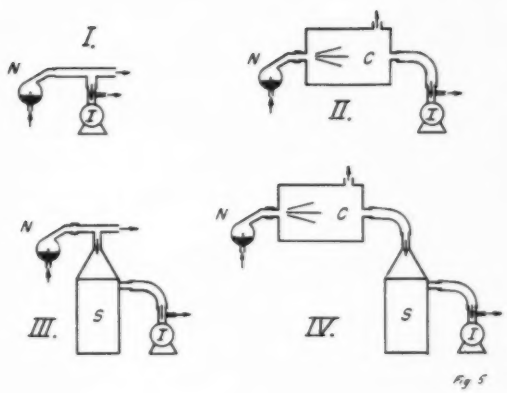


Fig. 5 Scheme of equipment sequence for generating and analysing aerosols (N, nebulizer; C, residence chamber; S, aerosol spectrometer; I, impactor).

In both cases care was taken that the flow from the nebulizer was substantially larger than the intake of the impactor ($F_n > F_i$) and that this excess would not cause a pressure variation of the flow.

The particulate components of the aerosol were eliminated prior to impaction, whenever required for control purposes, by passage through an aerosol spectrometer, or through a membrane filter. The latter has the disadvantage of requiring a pressure difference, also of exposing the gas to contact with an extended potentially sorbent surface.

Fig. 5 schematically indicates the combinations (I -IV) used for these tests. For testing aerosols, not generated by the nebulizer, the combinations (I) and (III) were employed (without N). -

- - - - - - - -

The apparent disadvantage of this mode of aerosol generation is - aside from the very short residence time - the contact of the future gaseous and particulate components in the same liquid, prior to aerosolization. This condition is easily overcome by the parallel use of several nebulizers, each containing one component only. This was not yet done, in order to study first this type of aerosol - generation, to be followed by the simultaneous separate dispersion of each component. Comparison between both, should give an insight into the combination rates between the same components in the gaseous phase.

- - - - - - - -

B.III. Quantitative Relations:

The following quantitative relations exist between the composition of the fluid in the nebulizer, the (irritant) gaseous and particulate components generated by it and the deposit densities resulting on the impacted surface. The symbols and units used are:

C_m = mass concentration of suspended or dissolved solids in stock solution (gr/cm^3);

C_v = volume concentration of gas in stock solution;

Q = volume of diluent added to 1 cm^3 stock solution (cm^3);

ϱ = density of solute (gr/cm^3);

d = particle diameter in colloidal suspensions (cm);

M = molecular weight of gaseous irritant;

W = ratio of nebulized (aqueous) liquid to total air flow (gr/liter);

$C_{(ppm)}$ = irritant (gas) concentration in air flow (ppm by volume);

M_A = mass-concentration of particulate matter in air flow (gr/liter);

N_A = number of particles in monodisperse aerosol (per liter);

F_i = flow rate through impactor (liter/min);

A_o = surface area exposed to impaction (cm^2);

t = length of exposure of (A_o) to impaction (min);

V_i = total irritant gas volume impacting surface (cm^3/cm^2)

M_i = total mass of particles impacting surface (gr/cm^2);

ν_i = number of statistical layers of impacted particulate matter on surface (cm^{-2}).

The constants characterizing the nebulizer and impactor operation in the tests described were as follows:
$W = 1.3 \cdot 10^{-2}$ gr/liter; $F_i = 6.8$ liter/min.; $A_o = 5.2$ cm^2;
hence $F_i/A_o = 1.31$ liter/cm^2/min.; $W \cdot F_i/A_o = 1.7 \cdot 10^{-2}$ gr/cm^2/min. -

- - - - - - - -

The concentration ($C_{(ppm)}$) of a gaseous trace component in the air flow, generated by nebulizing its aqueous solution, and the resulting impacted gas volume (V_i) are related as follows:

$$V_i = \frac{C_v \cdot W}{(Q+1)} \cdot \frac{F_i}{A_o} \cdot t = 2.5 \cdot 10^4 \cdot \frac{C_m \cdot W}{M(Q+1)} \cdot \frac{F_i}{A_o} \cdot t = C_{(ppm)} \cdot 10^{-3} \cdot \frac{F_i}{A_o} \cdot t \quad ..(1)$$

$$C_{(ppm)} = \frac{C_v \cdot W \cdot 10^3}{(Q+1)}; \text{ and: } Q = \left[2.5 \cdot 10^7 \cdot \frac{C_m \cdot W}{M \cdot C_{(ppm)}} \right] - 1 \quad .....(2)$$

Example:

To produce in the air flow a gas concentration of HCHO (M = 30), equivalent to 10 ppm from an aqueous solution (formalin, ($C_m = .36$), the nebulizer fluid has to consist of a dilution of 1 cm^3 in (Q = 390 cm^3 water) (eq.2). This air flow will cause on the impacted surface the volume contact of (eq.1):

$$V_i = 1.3 \cdot 10^{-2} \cdot t \text{ } cm^3 \text{ HCHO}/cm^2.$$

For soluble solids, the above relation can be modified to:

$$M_i = \frac{C_m \cdot W}{(Q+1)} \cdot \frac{F_i}{A_o} \cdot t = M_a \cdot \frac{F_i}{A_o} \cdot t \quad \ldots\ldots\ldots\ldots\ldots\ldots\ldots\ldots (1a)$$

$$\text{and: } M_a = \frac{C_m \cdot W}{(Q+1)} \ldots\ldots; \quad Q = \left[1.7 \cdot 10^{-2} \cdot \frac{C_m}{M_a} \right] - 1 \quad \ldots\ldots\ldots\ldots (2a)$$

Example:

To produce from a 10% stock solution ($C_m = 10^{-1}$) of NaCl an aerosol concentration ($M_a = 10$ mg/m^3 $= 10^{-5}$ gr/liter), 1 cm^3 of the solution has to be diluted with 130 cm^3 water (eq.2a) and results in an impact-density ($M_i = 1.3 \cdot 10^{-5} \cdot t$ gr/cm^2) (eq.1a).

- - - - - - - -

For suspensions of insoluble particles, e.g. spherical polystyrene latex particles of uniform particle diameter (d), the above can be further modified in terms of (N_a) and (ν_i) as:

$$\nu_i = 1.91 \cdot \frac{M_a}{\rho \cdot d} \cdot \frac{F_i}{A_o} \cdot t = 1.91 \cdot \frac{M_i}{\rho \cdot d} = N_a \cdot d^2 \cdot \frac{F_i}{A_o} \cdot t \quad \ldots\ldots (3)$$

$$\text{and: } N_a = 1.91 \cdot \frac{M_a}{\rho \cdot d^3} \ldots; \quad Q = \left[1.91 \cdot \frac{C_m \cdot W}{\nu_i \cdot \rho \cdot d} \cdot \frac{F_i}{A_o} \cdot t \right] - 1 \ldots (2b)$$

Example:

To produce an aerosol from a 10% hydrosol suspension of such latex particles ($\rho = 1$) of diameter ($d = 2 \cdot 10^{-5}$ cm $= .2$ μ) so that a statistical layer of ($\nu_i = 2$) results in ($t = 4$ min.), 1 cm^3 of the

hydrosol has to be diluted with ($\mathcal{V}$ = 324 cm^3 water) (eq.2b) resulting in an aerosol density of ($M_a = 4 \cdot 10^{-6}$ gr/liter) and ($N_a = 9.5 \cdot 10^8$/lit.) (eq.3). It follows that the impacted mass of the double layer is ($M_i = 2.1 \cdot 10^{-5}$ gr/cm^2), the number ($d^{-2} = 2.5 \cdot 10^9$/cm^2) with a total area of particle surface ($\pi \cdot \mathcal{V}_i = 6.3$), i.e. a more than 6-fold increase of surface area (and thus of the potential adsorption capacity); the total surface area of the (monodisperse) aerosol would be ($A_p = \pi \cdot N_A \cdot d^2 = 1.2$ cm^2/liter). -

- - - - - - - -

These relations have, however, certain limitations requiring critical application, particularly in case of the ratio (W) between liquid and gas issued by the nebulizer. If the liquid is volatile, its partial evaporation in the nebulizer will produce a gradual increase of concentration ($C_m/(Q+1)$), if the solute is less volatile (e.g. NaCl, Latex, etc.), and vice versa for gaseous solutes. This effect will cause (M_a) to be initially smaller, and larger after prolonged operation of the nebulizer than resulting from (eq.2a); it can be determined by individual calibration and minimized by frequent replacement of the nebulizer fluid. -

Furthermore, the operation of the nebulizer causes an increase of the partial pressure of the evaporating solvent in the aerosol, i.e. for water an increase in humidity. The value of ($W = 1.3 \cdot 10^{-2}$ gr/lit. for water) corresponds to a dew point at 15° C, i.e. to a relative humidity of ca. 53% at prevailing room temperature (27-28° C). This value is increased by the compressed air (rH<15%) fed into the nebulizer to ca. 65%, hence the temperature in the system may not be dropped below (ca. 20° C) in order to avoid condensation. -

- - - - - - - -

B.IV. BACTERIOLOGICAL PROCEDURE

1.) The Working Principle of the Concentrometer:

The method is based on the concentrometer[2] principle, whereby a graded concentrate of suspended particles is produced on a uniformly microporous filter matrix, by passing the suspension through a filter, which acts predominantly as a screen.

Contrary to the usual practice of the filtration whereby each surface element of the filter is passed by an equal fraction (V) of the total liquid volume, this fraction varies in the concentrometer along one direction (x) of a rectangular filterstrip in accordance with a predetermined mathematically defined function. Consequently, if the concentration within the liquid is uniform, the deposit on the screen will represent a "wedge", the thickness of which varies in a known way in the (x) direction. Equal quantities of fluid of different concentration will thus result in "wedges" of different thicknesses for corresponding positions on the screen.

The scheme of a concentrometer is represented in fig. 6 : the liquid is filled into a profiled cavity (A) of semi-circular horizontal cross-section (A) which is sealed by a plate (C) and by a resilient thin metal sheet (H). (H) has an exactly dimensioned rectangular window, which determines the area of the membrane available for filtration. This window coincides with a rigidly supported fine wire screen (M), which, in turn, supports the membrane strip, sandwiched between (H) and (C) during filtration. The plate (C) has a direct connection (E) with the cavity through which the liquid

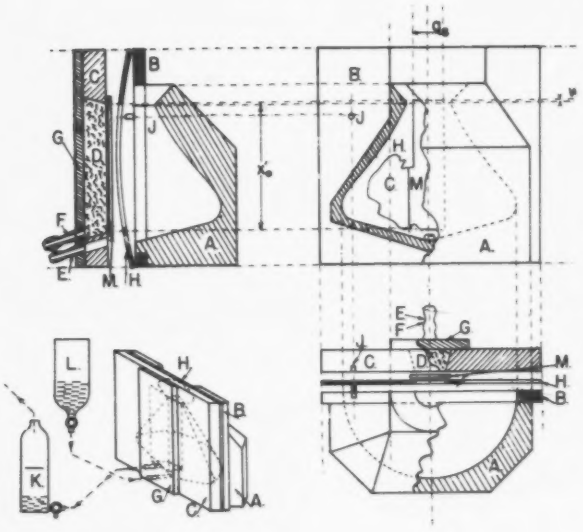


Fig. 6 Design of concentrometer for liquids: A, cavity block; B, flange plate of A; H, ph.-bronze gasket defining exact screen area; C, lucite screen holder, sealed by plate G; J, alignment pins (2); M, filter membrane; F, connects with vacuum flask K; E, with sample container L.

is filled initially from (L) until it overflows (A), after which (E) is closed. The second connection (F) leads to the back of the membrane, i.e. into (D) and is connected through the bottle (K) with a vacuum pump.

The pressure difference across (M), forces the liquid to flow from (A) through (M-D-F), while simultaneously the liquid level in (A) will be lowered. Consequently the fraction of (M), available to the passage of the liquid, will become steadily smaller and the lower parts of (M) must pass increasing quantities (V) in order to further lower the level in (A). When the level has reached the lower edge of (M), the fluid remaining in (A) is drained through (E), whereafter (C) is parted from (A) and (M) is removed. (M) carries on its surface the retained concentrate at a density varying along the vertical (x) direction in accordance with a function (V(x)) which is once and for all defined by the profile of the cavity (A).(+) Such a strip carrying concentrometric deposits is termed a "concentrogram" (C-gram).

(+) (V(x)) does not depend on the rate of filtration (i.e., pressure difference or flow-permeability of the screen material), and thus not on a gradual decrease of the flow rate, if partial clogging of (M) occurs. The sole requirement is the uniform porosity of (M) at the beginning of the operation.

The evaluation of such C-grams is determined by the following considerations: as the distribution of the flow density

through the screen along (x), (V(x)), is always the same, 2 different liquids (of similar nature of suspended matter) with the respective concentrations C_1 and C_2 produce from the same cavity 2 C-grams represented by:

$$C_1(x) = u \cdot V(x); \quad C_2(x) = w \cdot V(x); \quad \text{or} \quad \frac{C_1(x)}{C_2(x)} = \frac{u}{w} \ldots (1)$$

The general mathematical theory of the generation of these distribution functions is somewhat involved and will be published elsewhere.(3)

While it is physically possible to produce a large (but not infinite) variety of different distributions, V (x), by the suitable design of the cavity shape the selection of a logarithmic (slide rule) function:

$$V(x) = A \cdot e^{kx} \ldots (2)$$

where A, k are fixed instruments constants, is uniquely suitable for this particular purpose.

The principle underlying the evaluation of C-grams with a logarithmic density distribution is illustrated schematically in Fig. 7 where C_1 and C_2 stem from an unknown, C_s from a known (standard) concentration. It is evident that the shift of C_1 and C_2 against C_s by the distances d_1 and d_2 respectively, juxtaposes surface sections with identical variation of deposit density. This causes between the 2 C-grams a sharply defined position of equal appearance, demonstrated in Fig.7 by the precise matching of the lines, for their

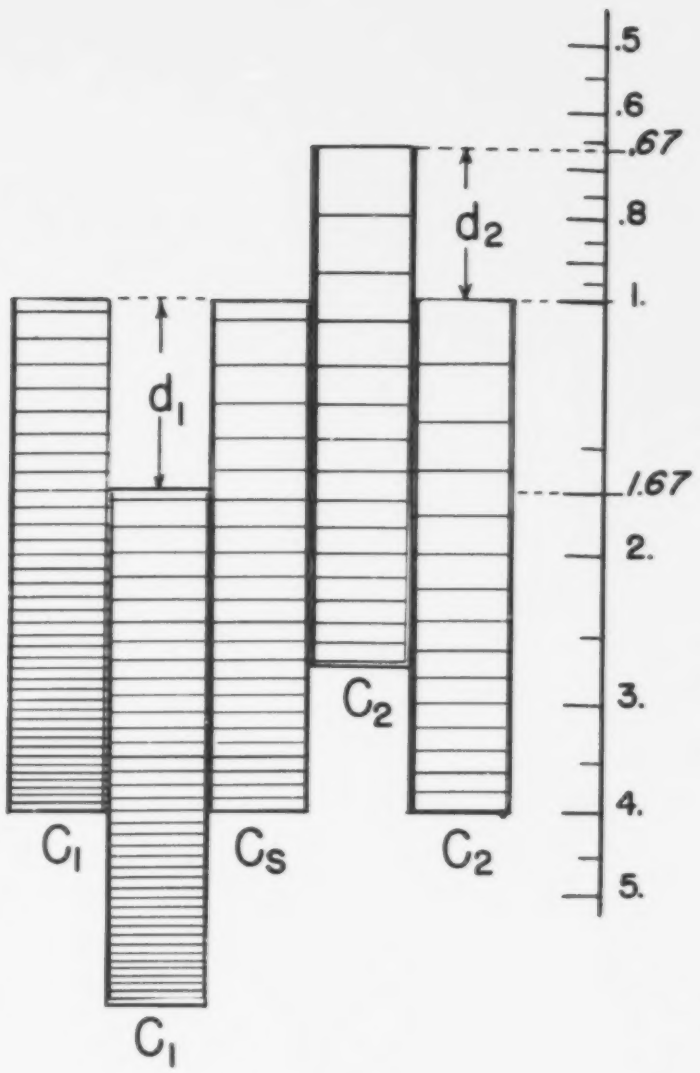


Fig. 7 Scheme of comparison of two (unknown) C-grams. (C_1 C_2) with a standard C-gram. (C_s) of known concentration. The concentrate density which increases downward according to a logarithmic function is represented by the closeness of the horizontal lines. Shifting of (C_1) (left) for the distance (d_1) against (C_s) produces an exact matching of the lines and analogously the shift of (C_2) (right) for (d_2). The distances (d_1, d_2) determined on the instrument scale (right), results in the concentration ration $C_1 : C_s : C_2$ = 1.67: 1 : 0.67, the total range in this example being 4.

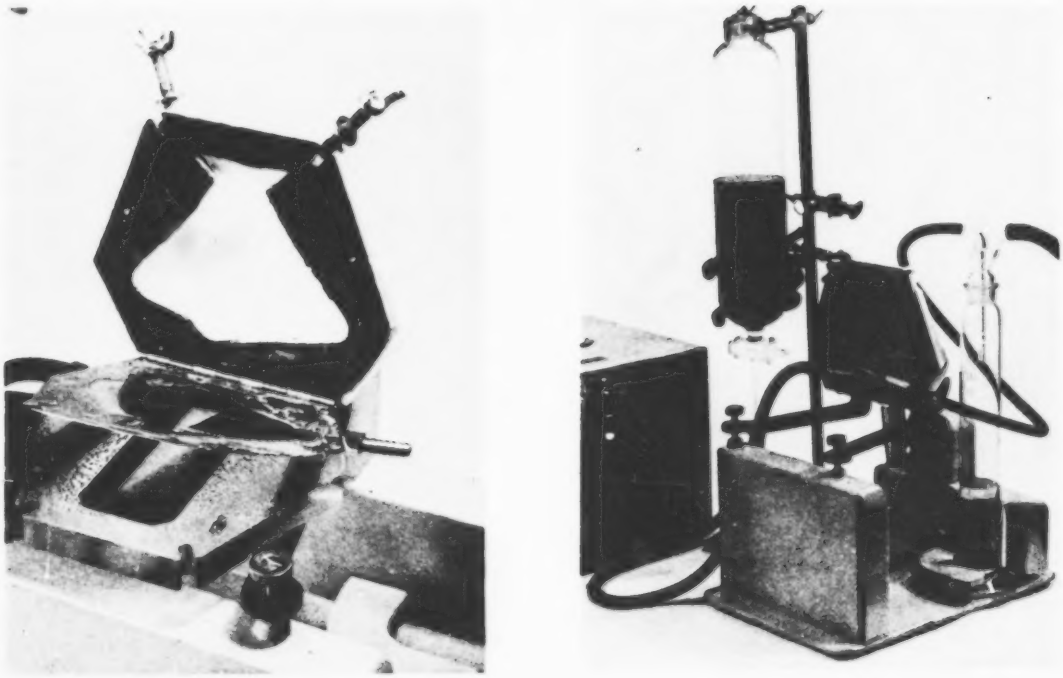


Fig. 8 Photograph of concentrometer assembly, (left) open; (right) during filtration.

density (closeness) corresponds to that of the deposit on the C-grams and varies along (x) according to eq. (2) and the scale in Fig. 7. In the example the shifts d_1 and d_2 correspond on the scale to:

$$C_1 = 1.67 \cdot C_s;\quad C_2 = .67 \cdot C_s;\ \text{or}\ C_1 : C_s : C_2 = 1 : 1.5 : 2.5$$

No other but the logarithmic variation of V(x) allows this simple mode of comparison, well-known in photometry. For the deposit density at any distance x_1 along x, eq. (1) results in:

$$\frac{C_1}{C_s} = \frac{e^{k(x1 + d)}}{e^k \cdot x_1};\quad \text{or}\quad \log \frac{C_1}{C_s} = k \cdot d;\quad \text{or}\quad \frac{C_1}{C_s} = \text{antilog}\ (k \cdot d)$$

i.e. the logarithm of the concentration ratio is independent of x_1 and directly proportional to the relative shift d. The total range, defined by the length of the screen in terms of the corresponding scale, represents in this example the factor of 4, which, for practical purposes, can be arranged to lie between $10^2 - 10^3$, the range depends on the shape and capacity of the cavity A.

Fig. 8 is a photograph of the instrument, used in the BIA-tests. The filter area is (2 cm x 8 cm) through which a total volume of 220 cm^3 are filtered, it has a range of 200, i.e. the volume which has passed the lowest horizontal strip of 2 mm width is 200 times larger than that which has passed the uppermost strip of equal width.

2.) Preparation of the bacterial I-strips

In almost all tests the same strain of E.coli was used because this organism is relatively insensitive to partial desiccation, while exposed in the impinger.

The procedure was as follows:

A 24-hour broth-culture (1 cm^3) was serially diluted in three 1:10 steps, 1 cm^3 of the last was diluted 1:100, and after thorough mixing diluted further to 1:1,000, resulting in an overall dilution of 10^{-6}. The diluent was a sterile .5% NaCl solution. - Each filtration with the concentrometer required 220 ml.

The concentrometer was sterilized by the incomplete combustion of ca. 1 ml Methanol (Formaldehyde) for 15 min,[1] each filtration required ca. 3 min. The membranes were then put on double strips of filter paper impregnated with a general nutrient solution, containing .1% Sodium Benzoate, and layed out in plastic dishes with air tight closures. The addition of the latter preserved substantially the viability of the deposit prior to insertion into the impactor.

In order to maintain nearly saturated humidity a wad of wet cotton was attached to the lid. This procedure was developed after it became evident that the time elapsed between deposit on the membrane and exposure to the aerosol was a significant factor in the subsequent growth development. During the exposure the membrane (I-strip) rested on a strip of filterpaper impregnated with dist. sterile water. After the exposure in the impactor, the membrane was removed and placed upon a rehydrated commercial nutrient

pad,(+) in the same plastic dish with cotton wad as before. After completion of the test series all dishes (each containing 3 I-strips) were incubated 20-24 hrs. at 37°C in vertical position, to avoid the formation of drops of condensation on the membrane surface.

After incubation the I-strips were stained with methylene blue by a technique, previously described by the senior author.(4)

A typical set of 4 microbiological C-grams varying in concentration as 1 : 5 : 25 : 125 is reproduced in fig. 9.

- - - - - - - - -

(+) Isopor DSN type "General I", supplier: AG Chemical Company, Inc. Pasadena, California

B.V. Results of (BIA) Tests.

While the principal effort was directed primarily toward the development of an optimal standardized procedure, a number of specific applications of the BIA test were explored during the period of the contract:

1.) Automobile Exhaust:

1.) In a previous report(5) the authors have described a series of tests on irradiated automobile exhaust, which demonstrated that the removal of the particulate component by the Aerosol Spectrometer could very substantially decrease the eye irritation recorded by a calibrated human panel.

Two similar tests at the identical installation(+) with a 30-40% olefine containing (i.e. aerosol forming) fuel of high irritant capacity were conducted. In these BIA tests the irradiated exhaust was fed to the impactor directly, and after passage through either a membrane filter, or the aerosol spectrometer.

(+) Stanford Research Institute, South Pasadena, Calif., courtesy of Air Pollution Foundation, San Marino, Calif.

Fig.10 (I, II) is a reproduction of the so resulting I-strips. The presence of the aerosol produced a clearly defined track of growth inhibition persisting either through part of, or the full length of the I-strip. Since considerably different bacterial concentrations were used in both tests, a shift (s.a.) about a

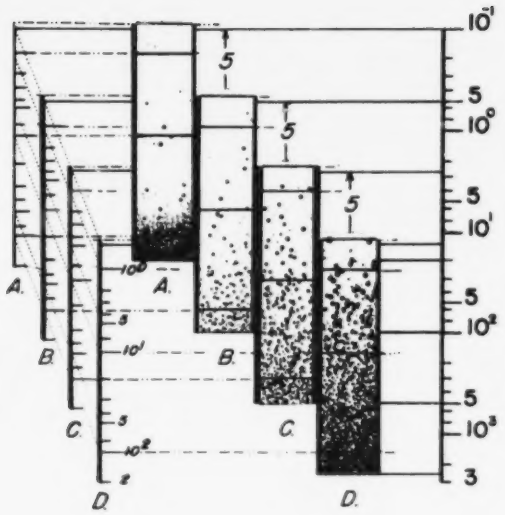


Fig. 9 Photo of 4 C-grams, carrying bacterial concentrations, increasing by factor 5; (on right) logarithmic concentrometer scale, corresponding to the cavity profile used.

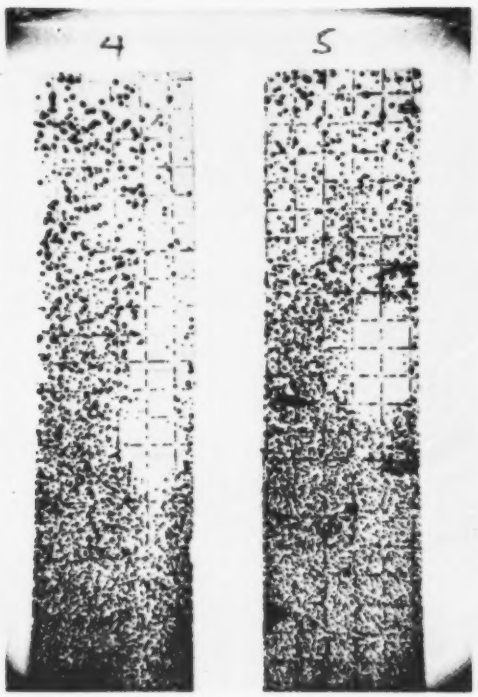
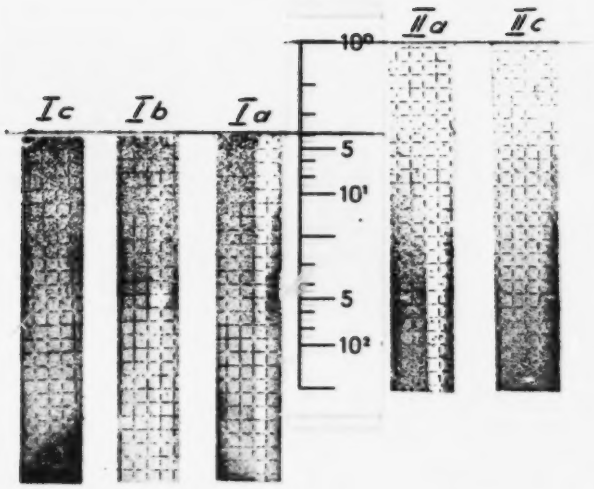


Fig. 11 A record of tobacco smoke aerosols on I-strips, producing topic growth inhibitions.



Test Ia, IIa	irradiated exhaust	direct	
Test IIb		dto.	through membrane filter
Test Ic, IIc		dto.	through aerosol spectrometer

Note: The local growth inhibition (white spots) on Ia and Ib are due to temporary (accidental) interruption of the rotor motion and represent local regions of largely increased exposure to impaction.

FIG. 10

Fig. 10 I-strips showing reduction of auto exhaust inhibition through membrane filter and aerosol spectrometer.

factor 4 is necessary to render the 2 tests comparable.

Although at present too few such tests are available to attach to them more than a qualitative significance, it is apparent that passage of the aerosol through a membrane substantially decreased - and passage through the aerosol spectrometer entirely eliminated the growth inhibition. This indicates that the inhibitory agent is associated with particulate matter of a size which can be eliminated by the application of centrifugal forces of ca. 25,000 g , i.e. of Stokes' diameters $[0.3\ \mu \geq d \geq .08\ \mu]$. -

- - - - - - - -

2.) Tobacco Smoke:

Although only indirectly connected with the specific problem, a number of BIA records were taken directly from the (air-conditioned) laboratory atmosphere (t = 12 min). During the test a burning cigarette was brought several times for a few seconds near the inlet of the impactor. This resulted in a typical temporary irritation record coincident with the impacted traces of tar-aerosols visible on the I-strips (fig. 11).

This test indicates the very large sensitivity to even minute aerosol concentrations - also the lack of parallelism between the BIA-test and the reactivity of the human for this type of irritation.

- - - - - - - -

3.) Synthetic Aerosols:

In order to explore the possibility of reproducing the synergistic irritation recorded in animals, by LaBelle et.al.[6] and particularly by Amdur (see Appendix), synthetic aerosols containing HCHO as irritant were generated as described above. As particulate component was used either KCl or a monodisperse suspension of polystyrene latex of (d = 0.26 μ), in order to compare the effect of a water soluble salt of a limited affinity for HCHO with a probably entirely neutral particle substance. -

Both, irritant and aerosol concentrations, were of the same order of magnitude as those employed in the animal experiments, namely:

HCHO ..... $C_{ppm} = 30 \sim V_i = 4 \cdot 10^{-2} \cdot t$ (eq.1a)
KCl ..... $M_a = 38$ mg/m$^3 \sim M_i = 2 \cdot 10^{-4}$ gr/cm^2 (eq.4a)
Latex ..... $M_a = 53$ mg/m$^3 \sim (\nu_i = 10)$ (eq.5, 6)
Latex ..... $M_a = 16$ mg/m$^3 \sim (\nu_i = 3)$ (eq.5, 6)

The test pattern consists of the following combinations:

#	HCHO	KCl	Latex
1	-	-	-
2	-	x	-
3	-	-	x
4	x	-	-
5	x	x	-
6	x	-	x

In addition parallel tests were conducted with potentially synergistic combinations (#5, 6) by passing the aerosol through an aerosol spectrometer, prior to its entry into the impactor.

The equipment set-up corresponded to fig. 5 II and IV respectively, hence the "age" of the aerosol was 45" - 1 min., i.e. closely similar to that in Amdur's work (see Appendix) for the combination HCHO-NaCl. Fig.12 A and B-C represent photographic reproductions of 2 series of I-strips, exposed to synthetic aerosols and selected to illustrate the following.(+)

(+) The sets differ slightly with regard to bacterial concentration and impaction rate. Within each set all I-strips are identical.

The results so far available, can be summarized as follows:

a.) The plain control (#1) proved unnecessary since the not impacted side of each strip gave almost always sufficient evidence of normal growth.

b.) The KCl control (#2) produced no traceable effect.

c.) Heavy latex deposits (#3) - as control - (ν>10) proved partially inhibitory. This effect can possibly be attributed to the covering of the cells with latex layers, causing a physical restriction of oxygen access. However, a (minor) specific inhibitory action of the latex as such is not quite disproven at present.

d.) A definite effect is produced by HCHO (#4) at the

concentration used, which represents the contact of irritant gas of $\left[V_i = 1.7 \cdot 10^{-1}\ cm^3/cm^2\right.$ for t = 4 min, and $V_i = 1.9 \cdot 10^{-1}\ cm^3/cm^2$ for t = 12 min$\left.\right]$. -

Characteristic for the irritant without aerosol, is the absence of a discrete path, instead of which a more general diffuse growth inhibition occurs. This may be attributed to the effect of gas diffusion: (A:a vs. b)

e.) The coexistence of the irritant with KCl (#5) produced a clearly defined path of inhibition (A:c, B:b,c), strongly indicative of a synergism, particularly because comparison of (#4) with (#5) shows better growth development for the area not impacted and much less for the path of impaction. (Fig.12 A:b vs. c)

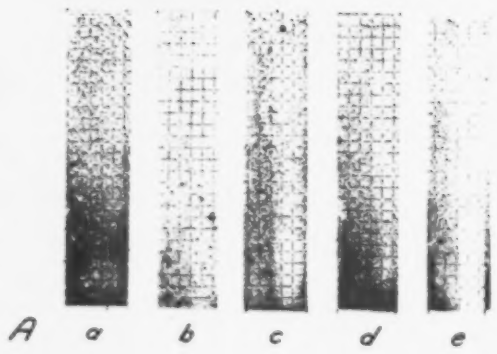
This can be interpreted as being due to an accumulation of irritant on the aerosol, which should cause not only a topical intensification at the locus of impaction, but also a depletion of the gaseous component available for diffusion.

f.) The coexistence of irritant with latex (#6) resulted a pattern similar to (#5), however, to a much larger extent. (Fig.12 A:c vs. e).

As for the exhaust aerosols passage through the aerosol spectrometer - operated at (N = 18,000, O = 40) - eliminated all particles of ($d \geq .2\ \mu$) and the major fraction of those below this size.

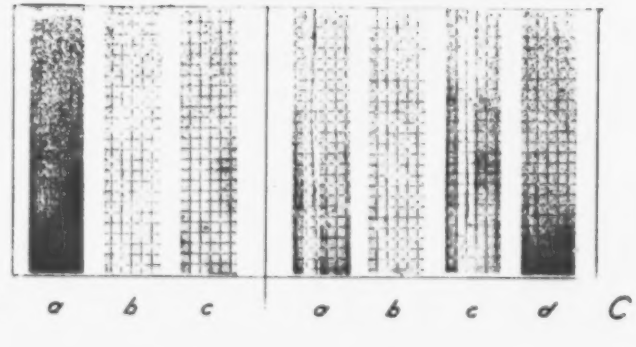
If part, or all of irritant molecules were associated with the aerosol particles, the inhibition resulting from the exhaust should be less or entirely absent. This pattern was apparent in the

Fig 12A



	Date	HCHO $V_1\left[cm^3/cm^2\right]$	KCl $M_1\left[gr/cm^2\right]$	Latex ν_1	t (min)	Centrifuge
A	7/22/58				12	
a	"	-	-	-	12	-
b	"	$1.9 \cdot 10^{-1}$	-	-	12	-
c	"	$1.9 \cdot 10^{-1}$	$4 \cdot 10^{-4}$	-	12	-
d	"	$1.9 \cdot 10^{-1}$	$4 \cdot 10^{-4}$	-	12	x
e	"	$1.9 \cdot 10^{-1}$	-	1.3	12	-

Fig 12 A,C



	Date	HCHO $V_1\left[cm^3/cm^2\right]$	KCl $M_1\left[gr/cm^2\right]$	Latex ν_1	t (min)	Centrifuge
B	7/29/58				4	-
a	"	$6.7 \cdot 10^{-3}$	$1.4 \cdot 10^{-4}$	-	4	-
b	"	$1.7 \cdot 10^{-1}$	$7 \cdot 10^{-5}$	-	4	-
c	"	$1.7 \cdot 10^{-1}$	$1.4 \cdot 10^{-4}$	-	4	-
C	"				4	
a	"	$6.7 \cdot 10^{-3}$	-	1.3	4	-
b	"	$1.7 \cdot 10^{-1}$	-	.65	4	-
c	"	$1.7 \cdot 10^{-1}$	-	1.3	4	-
d	"	$1.7 \cdot 10^{-1}$	-	1.3	4	x

Fig. 12 (A, B, C) I-strips showing synergistic effect of HCHO and particles of KCl or latex, also the effects of each separately.

tests, more specifically: Passage through the aerosol spectrometer by the

g.) HCHO-KCl aerosol diminished, but did not entirely eliminate its inhibitory effect. (Fig. 12 A:c vs. d)

h.) HCHO latex aerosol eliminated its inhibitory effect completely. (C:a,b,c, vs. d)

This difference can have three causes:

α.) incomplete elimination of KCl, because of the (undefined) very small particle size;

β.) evaporation of HCHO from the KCl particles on the wall of the (not cooled) aerosol spectrometer;

γ.) a lesser surface capacity of KCl compared with latex;

δ.) a larger concentration of HCHO than capable of association with KCl.

In order to explore these alternatives, several test series were made, where the relative concentration between HCHO and aerosol were changed, by using $C_{(ppm)} = 32$ or $= 1.3$, i.e. the ratio particle surface/gas was increased by a factor 25; also for $C_{(ppm)} = 32$ one half of the aerosol concentration (M_a) was used, i.e. the ratio was decreased by a factor 2 for the same partial pressure (p) of the irritant:

For KCl the effect increases markedly with (p) for the same (M_a) (B:a vs. b,c). For the same (p) and different (M_a), the inhibitory effect appears unchanged, though the "side"-growth (on the not-impacted area) heavier. (B:b vs. c). This coincides

with the pattern observed for the irritant with and without KCl (A:b vs. c) and seems to lend support to the interpretation by diffusion effects in the not impacted area by "free" HCHO, which for the same (p) would be the larger the smaller (M_a).

The pattern is somewhat different for latex: The inhibition is much more marked than for KCl and appears almost independent of concentration of the irritant (C:a vs. b,c), though dependent on (M_a) (C:b vs. c). However, the "side-growth" is distinctly larger for large values of (M_a) (C:a,c vs. b). This agrees with the observation for KCl and indicates also that the associative capacity for HCHO by latex is larger than by KCl - if commensurable particle surface area, i.e. diameters, can be assumed.

Regarding the above alternatives ($\alpha - \delta$) these tests, - though strictly exploratory - support (γ) and (δ) and do not require (α). -

Clarification of these alternative explanations necessitates extended additional experimentation.

- - - - - - - -

Conclusion:

The BIA-test - as developed at present - is able to indicate qualitatively and to some extent quantitatively the irritation intensity (smog, tobacco smoke, HCHO aerosols) of aerosols. It has been used in synthetic aerosols as indicator for the degree of association of the irritant gas (formaldehyde) with air-borne particulate matter, by comparative tests, prior to and after passage through the aerosol

spectrometer.

The findings support the assumption that a fraction of the irritant gas molecules associates with the particle surface, and that this association produces an intensifying (synergistic) effect - which increases with the aerosol concentration.

The synergistic effect parallels the pattern of Amdur's tests with guinea pigs, and apparently also those of LaBelle with regard to paraffin aerosols in analogy with latex dispersions.(6)

- - - - - - - -

ACKNOWLEDGEMENT

The work was performed under a Research Contract with the US Public Health Service, which also made the cooperation with Mr. Heber J. R. Stevenson of the Community Air Pollution Program possible by assigning him for this purpose to the California Institute of Technology.

The Los Angeles Air Pollution Control District through its research director, Dr. Leslie A. Chambers and members of his staff aided the work, also the Air Pollution Foundation by facilitating on several occasions access to the facilities at Stanford Research Institute, South Pasadena.

The AG Chemical Company assisted the work substantially by permitting the liberal use of its bacteriological facilities and the Instrument Development & Mfg. Comp. by contributing an Aerosol Spectrometer (Model E) as loan.

Mr. M. L. Warrick, Jr. has applied his mechanical skill to the construction of the impactors and various other experimental devices, Mrs. L. M. Hauck contributed her conscientious technical and clerical assistance.

- - - - - - - -

Subtexts to Fig. 1 - 12

Fig. 1 Vertical cross-sections and partial cuts of Rotary Impactor.

Fig. 2 Photograph of Rotary Impactor.

Fig. 3 Lucite holder for mounting I-strip from monodisperse latex aerosols (d = .26 μ).

Fig. 4 Rotary Impactor I-strip with impactor patterns: a.), for continuous (left) and intervalled (right) aerosol in flow (note sharp definition); b.), impaction patterns on stationary rotor (right) and analogous pattern on conventional slide impactor.

Fig. 5 Scheme of equipment sequence for generating and analysing aerosols (N, nebulizer; C, residence chamber; S, aerosol spectrometer; I, impactor).

Fig. 6 Design of concentrometer for liquids: A, cavity block; B, flange plate of A; H, ph.-bronze gasket defining exact screen area; C, lucite screen holder , sealed by plate G; J, alignment pins (2); M, filter membrane; F, connects with vacuum flask K; E, with sample container L.

Fig. 7 Scheme of comparison of two (unknown) C-grams. (C_1 C_2) with a standard C-gram. (C_s) of known concentration. The concentrate density which increases downward according to a logarithmic function is represented by the closeness of the horizontal lines. Shifting of (C_1) (left) for the distance (d_1) against (C_s) produces an exact matching of the lines and analogously the shift of (C_2) (right) for (d_2). The

distances (d_1, d_2) determined on the instrument scale (right), results in the concentration ration $C_1 : C_s : C_2 = 1.67 : 1 : 0.67$, the total range in this example being 4.

Fig. 8 Photograph of concentrometer assembly, (left) open; (right) during filtration.

Fig. 9 Photo of 4 C-grams, carrying bacterial concentrations, increasing by factor 5; (on right) logarithmic concentrometer scale, corresponding to the cavity profile used.

Fig. 10 I-strips showing reduction of auto exhaust inhibition through membrane filter and aerosol spectrometer.

Fig. 11 A record of tobacco smoke aerosols on I-strips, producing topic growth inhibition.

Fig. 12 (A, B, C) I-strips showing synergistic effect of HCHO and particles of KCl or latex, also the effects of each separately.

BIBLIOGRAPHY

1.) Task Group Report. Technique of Bacterial Examination of Water with Molecular Filter Membranes. Journal AWWA, 45, 1196-1210, 1953.

2.) A. Goetz. The Application of the Concentrometer Principle to the Determination of Aerosols. Journ. APCA, 6, 72-76, 1956

3.) A. Goetz. Theory, Methodology and Application of the Concentrometer, Industr. Assoc. Calif. Inst. of Tech., (Aug.1955)

4.) A. Goetz and N. Tsuneishi. Application of Molecular Filter Membranes to the Bacteriological Analysis of Water. Journ. of AWWA, 43, 943-969, 1951.

5.) A. Goetz and H.J.R. Stevenson. The Aerosol Spectrometer Its Theory, Construction and Application to the Analysis of Exhaust and Atmospheric Aerosols. Proc. APCA Meeting San Francisco, Nov. 1957.

6.) LaBelle, Charles W.; Long, James E.; and Christofano, Emil E. Synergistic Effects of Aerosols. AMA Arch. of Ind. Health, 11, 297-304, 1955.

- - - - - - - -

A P P E N D I X

INTERPRETATION OF THE DATA, REGARDING THE RESPIRATORY REACTION OF GUINEA PIGS TOWARD HCHO, IN THE PRESENCE AND ABSENCE OF NaCl AEROSOLS

ABSTRACT

A quantitative analysis of the results obtained by Amdur(+) on the above subject is given whereby the relationship between partial pressure of irritant and particle surface area in the aerosol is discussed from which the degree of synergistic intensification relative to irritant and aerosol concentration is derived. The test data available for HCHO confirm this prediction over a wide range and indicate the statistical accumulation of 2 - 3 molecular layers on the surface of the NaCl particles. -

(+) Mary O. Amdur, Harvard School of Public Health, Harvard University, Boston, Mass., Progress Report February 1958, USPHS Research Grant S-8 and unpublished recent data by this author.

- - - - - - - -

This exposé is included in the Report as it represents an activity during the period of the contract - mainly, however, because it is closely related to the general subject matter of the contract.

- - - - - - - -

[Note: In the following, reference is made to: "An Interpretation of the Synergistic Effect of Aerosols, based upon Specific Surface-Action of the Air-borne Particles" by A. Goetz (still unpublished manuscript), wherein the synergistic effect of aerosols with gaseous trace components of irritants in the atmosphere is interpreted by the statistical accumulation of the latter onto the surface of the aerosol particles.]

- - - - - - - -

If the synergistic effect is due to surface accumulation of all of the irritant gas molecules (or fraction thereof) on the particle surface, it follows that:

a.) The number (per unit volume) of irritant molecules (N_m'), so accumulated, must be proportional to the total particle surface area (A_p) available.

b.) The toxicity increment, ΔT (= $T_s - T_g$) [where (T_g) the toxicity in terms of the physiological signal of irritation for the gaseous irritant alone; and (T_s) the corresponding toxicity in the presence of the aerosol] must be dependent only on the aerosol concentration (C_s) and independent of the concentration of the irritant gas (C_g).

c.) Condition b.) should be valid only in the range of low (C_g) values, where (T_s) is significantly larger than (T_g).

d.) For larger values of (C_g) the synergistic effect should disappear. ($T_s = T_g$)

e.) Condition d.) should be true only if the irritant molecules do not chemically react with, or diffuse into the particle. If this is the case and the resulting reaction product is less toxic, the intensivation by the aerosol ($T_s > T_g$) should revert to an attenuation ($T_s < T_g$).

In quantitative terms the following relations are significant: Since the NaCl-aerosols were prepared under nearly identical conditions, it can be assumed that the particle size (resp. distribution) is uniform and similar throughout, i.e. the mass-concentration of NaCl (C_s) reported, is proportional to the area (A_p) available.

For a (monodisperse and spherical) NaCl-aerosol of the particle diameter (d, in cm), the number of particles (N_p, per m^3) is for 1 mg (per m^3): $N_p = (10 \cdot d)^{-3}$, and their combined surface: $A_p = 3 \cdot 10^{-3} \cdot d^{-1}$, (An error in the determination of the actual d-value affects thus (A_p) reciprocally.)

The number of irritant gas molecules (per m^3) amounts for (C_g = 1 ppm) to: ($N_m = 2 \cdot 10^{19}$). If the area required for occupation by one molecule (either HCHO or HCOOH) is estimated in accordance with available data to: (15 $Å^2 = 1.5 \cdot 10^{-15}$ cm^2), the area required by one layer of molecules for (C_g = 1 ppm) amounts to ($A_m = 1.5 \cdot 10^{-15} \cdot 2 \cdot 10^{19} = 3 \cdot 10^4$ cm^2 per m^3). Hence the ratio ($n \cdot A_p/A_m = n \cdot d^{-1} \cdot 10^{-7}$) represents (per mg NaCl and ppm irritant)

the fraction (N'_m/N_m) of the irritant molecules present, which can be bound in (n) layers to the surface and thus produce the synergistic effect.

Hence the "synergistic capacity" ($n \cdot A_p/A_m = \Sigma$) of an aerosol should be the larger, the smaller (d), and the larger (n), i.e. the number of layers which can exist statistically on (A_p). If (d) is known, (n) can be established from the variation of (ΔT) with (C_g) for constant (C_s) (c.f.). It is to be expected that (n) increases exponentially with decreasing temperature - (hence (Σ) (i.e. the potential toxicity) of a suitable irritant-aerosol system should increase sharply with a drop of temperature!). -

Amdur estimated from electron microscope determinations $d = 4 \cdot 10^{-6}$ cm, (the writer expects the actual size, due to the hygroscopic nature of NaCl noticeable already at rH = 50%, to be more likely $5\text{-}6 \cdot 10^{-6}$ cm). For ($d = 4 \cdot 10^{-6}$ cm) ($A_p = 750$ cm^2) per mg NaCl. Three (average) levels of C_s (= 3 mg, 10 mg, 30 mg) were used corresponding to respective A_p-values of: ($2.25 \cdot 10^3$, $7.5 \cdot 10^3$, $2.25 \cdot 10^4$ cm^2).

The C_g-values were in the range ($.2$ ppm $\leq C_g \leq 50$ ppm) for both irritants, corresponding to $[6 \cdot 10^3 \leq A_m \leq 1.5 \cdot 10^6]$. Obviously, the lower C_g-range results in values of (A_m), which are of the same order of magnitude as the A_p-values, resulting from the C_s-range employed. It can thus be predicted that the synergistic effect ($\sim \Delta T$) will become apparent in the lower range of (C_g), i.e. where ($n \cdot A_p$) approaches the value of (A_m).

The absolute value of ($n \cdot A_p$) is, however, at present not known exactly - it depends on (d) and on the value of (n), i.e.

the number of irritant molecules which can be carried per unit particle surface. The uncertainty of (Σ) depends thus on (n/d). Under the same conditions of temperature, humidity and "age" of the irritant-aerosol system, (n) should be independent of (d), (C_s) and also of (C_g) - provided the irritant does not react with the particle substance.

If (d) is approximately known, (n) can be evaluated from (ΔT). The so resulting value of (n) should be small ($1 < n < 10$) in accordance with existing experience with statistical surface layers.

It is considered very fortunate that Amdur's investigations for the first time supply all data required for this type of quantitative evaluation, and a physiological irritation signal, which appears approximately free of higher-order complexities.

The data were treated as follows:

The variation of (T_g) with (C_g) was established as a continuous function (though more data would be very desirable).(fig.1)

The ratio ($A_m/A_p \sim 1/\Sigma$) was established, according to the above, from the data of (C_g and C_s) for ($d = 5 \cdot 10^{-6}$ cm), i.e. ($A_m/A_p = 10^7 \cdot d \cdot C_g/C_s = 50\ C_g/C_s$).

In fig. 2 (A_m/A_p) is plotted vs.(T_g),(T_s) and (ΔT) for HCHO and HCOOH.

Both irritants show an initial rise of (ΔT) to and (for HCHO) constancy beyond (A_m/A_p = 2-3). From the above follows that ($n \sim 2$-3), i.e. (A_p) is "saturated" by 2-3 (statistical) layers of irritant molecules. If (C_g) is sufficient to produce this saturation (ΔT) is maximal and does not increase even <u>if (C_g) and thereby (A_m) is increased 100-fold.</u>

It is noteworthy and a fair demonstration of the hygienic significance of the phenomenon, that at the saturation point - where (A_m/A_p = n) - the (then maximal) synergistic effect produces a toxicity (T_s), which corresponds (in the absence of the aerosol) to a 15-fold higher irritant concentration.

In order to test the validity of condition a) the writer suggested additional tests (for HCHO only) in the same C_g-range but for larger and smaller C_s-values. The resulting data are plotted (fig. 3) and appear to confirm the expectation, especially as the constancy of ΔT occurs for the same value of A_m/A_p = n and appears to be independent of (C_s).

The relation between (ΔT) and (C_s, resp. A_p) is - as would be expected - not linear, but approximately logarithmic [i.e. $\frac{\Delta T}{\lg C_s}$ = const] for ($A_m/A_p \geq n$), i.e. for "saturated" particles.

- - - - - - - -

A final remark may concern the question, whether or not this interpretation of surface accumulation of the irritant on (suitable) aerosol particles is the only one which meets the available data:

One alternative may be based on the assumption that the irritant would form a compound with NaCl, more toxic than the irritant itself: In this case (ΔT) should increase with both, (C_s) and (C_g) - as the concentration of this compound should depend upon both components.

Another alternative could be a dissolution of the irritant

within the particle. In this case one would anticipate a limited (A_m/A_p)-range where in (ΔT) is constant, because, as the irritant gas penetrates into the particle, it becomes less available - i.e. one would obtain a pattern similar to that shown for HCOOH - known to react with NaCl. -

This may be the reason why the absolute level of (ΔT), obtained for this compound, is but one third of that for HCHO, inspite of its substantially higher gaseous toxicity.

A further, seemingly very important difference is the relation of the condensation points of these irritants to room- resp. body temperature: ($-21^{\circ}C$ for HCHO and $+100.7^{\circ}C$ for HCOOH) which for the same (C_g) - i.e. same partial pressure - render the latter much more condensable than the former. Hence the 2 substances cannot be expected to form similar associations with the particle surface.

As already indicated, the "age" of the irritant-aerosol system should influence (ΔT) significantly - if the time interval between its generation and application is insufficient to establish an equilibrium state of the association between gas and particle surface. In Amdur's experiments the "age" of the system was apparently less than 1 minute, when it reached the test animals, - enough for rendering the effects of gas-diffusion negligable, since the average particle distance in the aerosol is very small (it varies for the C_s-values used between 15 - 35 micron), but probably much too short for establishing diffusion or reaction equilibria within the particle itself. -

- - - - - - - -

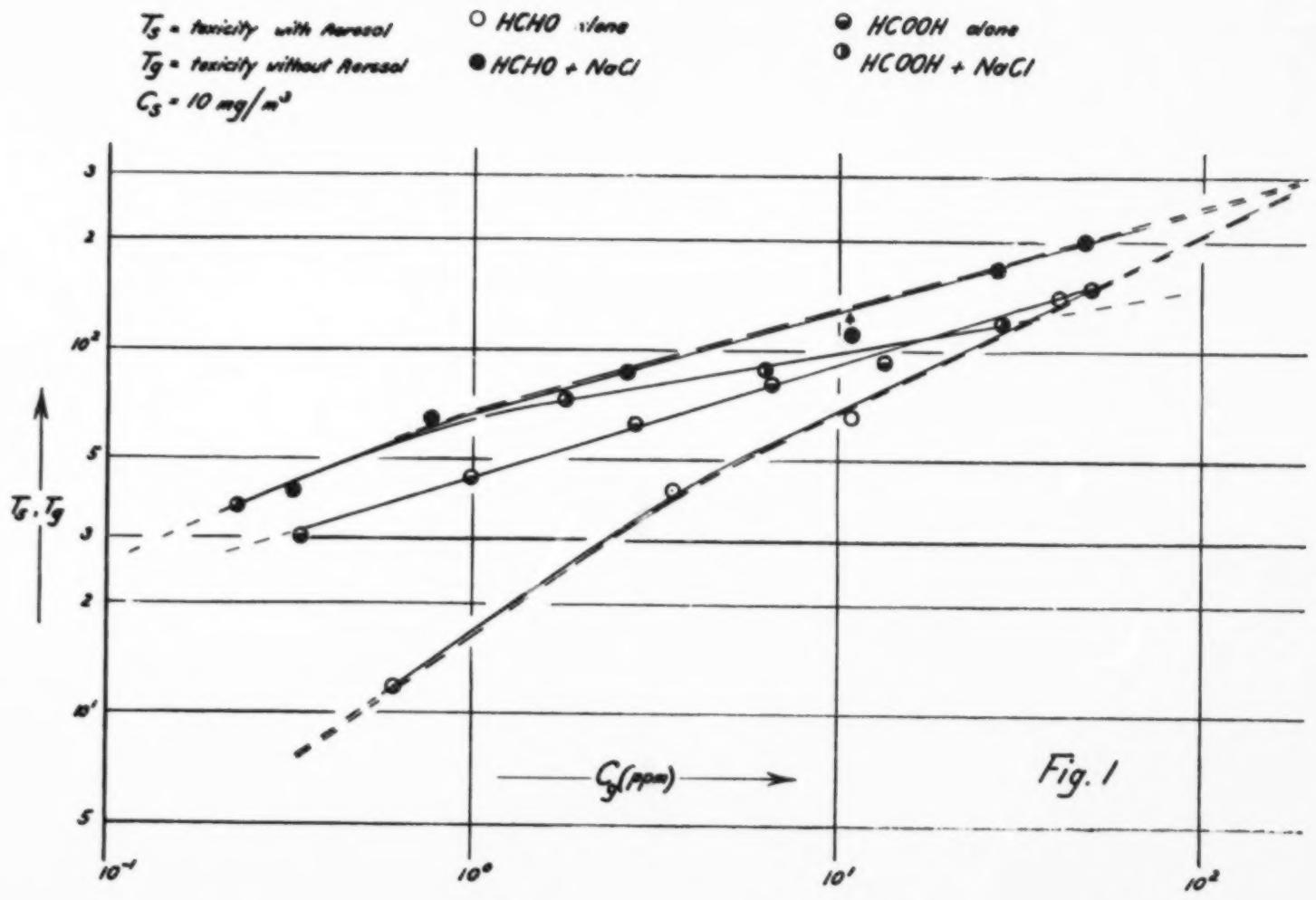

T_s = toxicity with Aerosol
T_g = toxicity without Aerosol
C_s = 10 mg/m³
HCHO alone
HCHO + NaCl
HCOOH alone
HCOOH + NaCl
$T_s : T_g$
C_g (ppm)
3
2
10^2
5
3
2
10^1
5
10^{-1}
10^0
10^1
10^2

Fig. 1

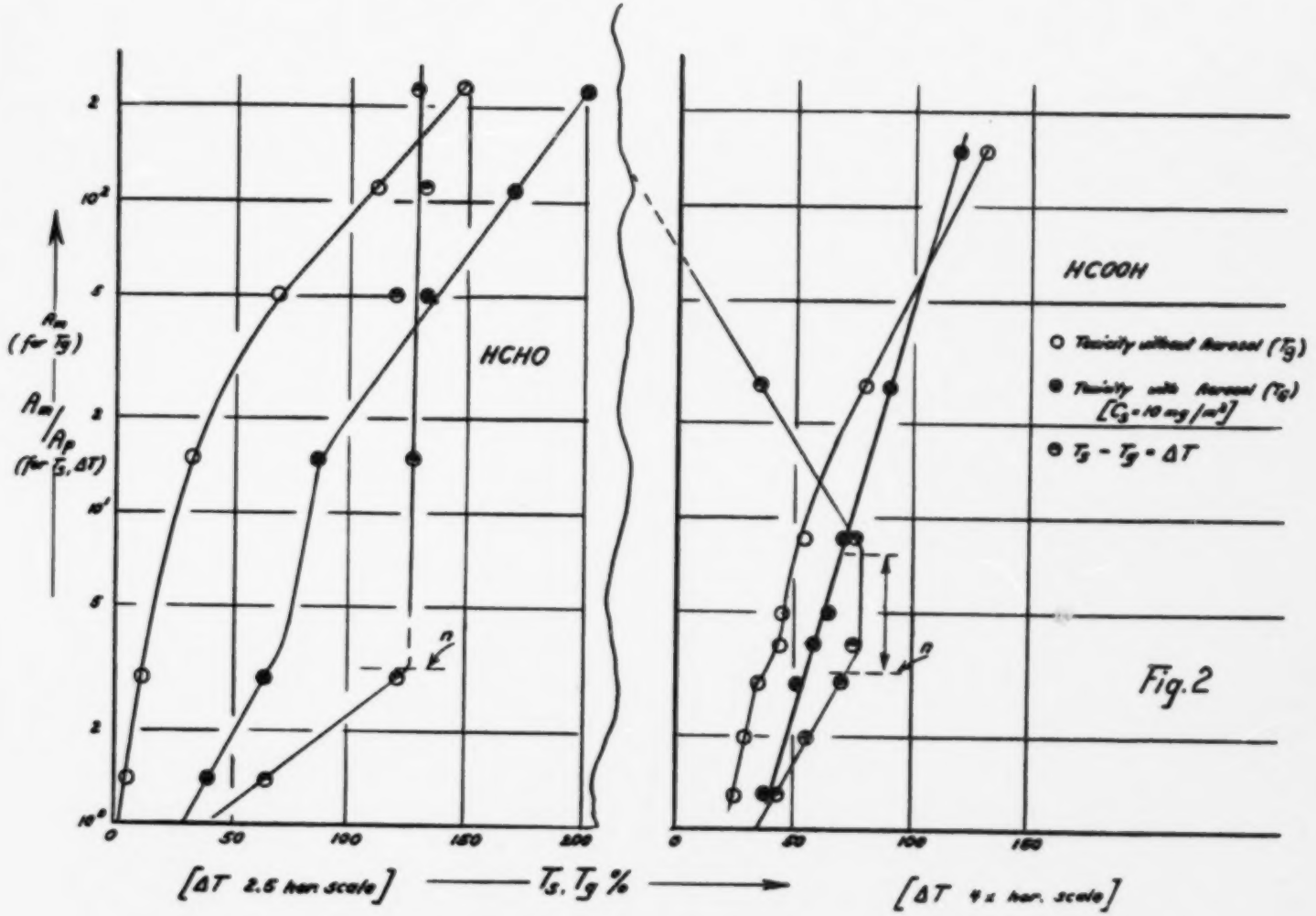

HCHO
HCOOH
Toxicity without Aerosol (T_g)
Toxicity with Aerosol (T_s) [C_s = 10 mg/m³]
$T_s - T_g = \Delta T$
A_m (for T_g)
A_m / A_p (for $T_s, \Delta T$)
[ΔT 2.5 hor. scale]
T_s, T_g %
[ΔT 4 x hor. scale]

Fig. 2

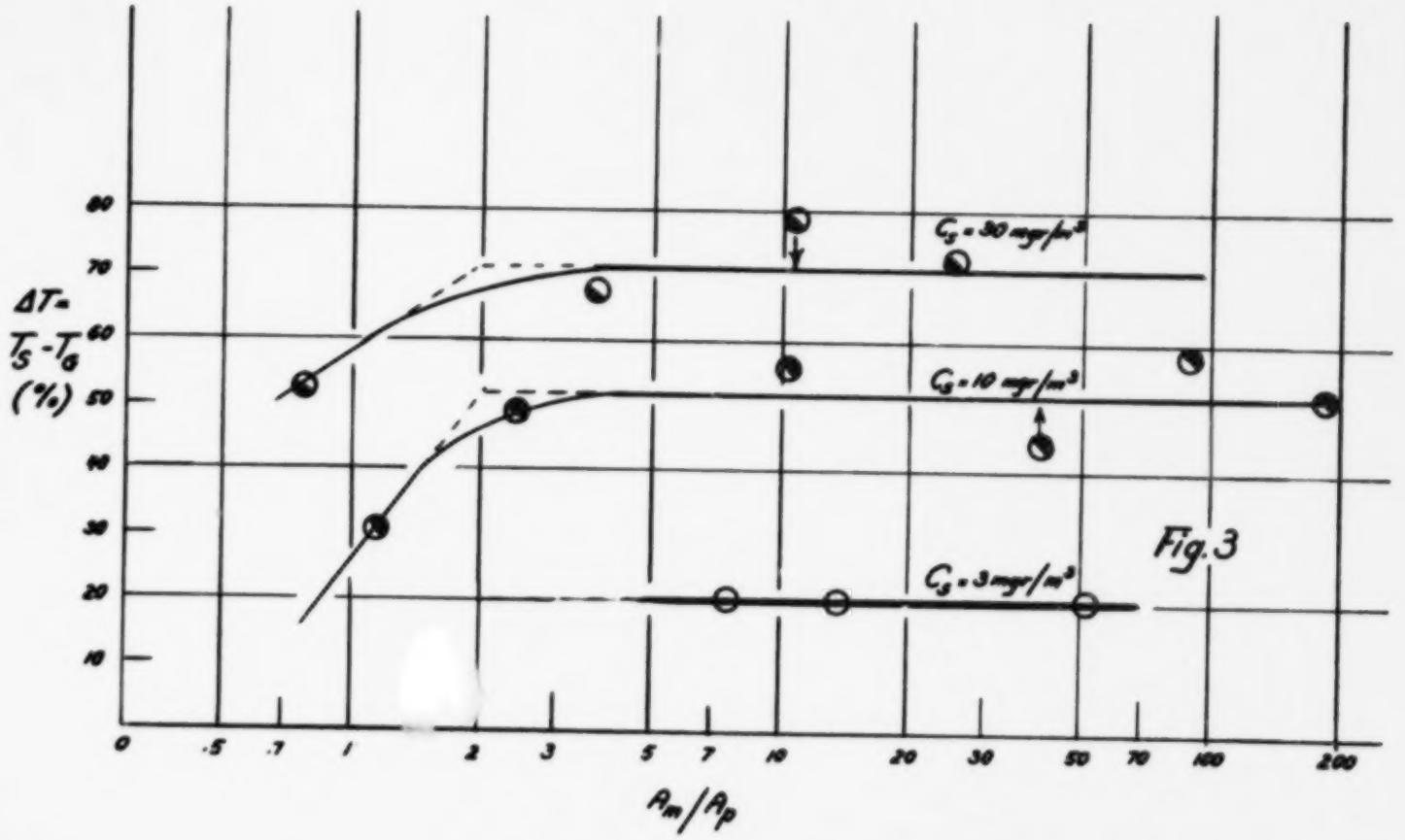

ΔT=
$T_S - T_G$
(%)
80
70
60
50
40
30
20
10
0
.5
.7
1
2
3
5
7
10
20
30
50
70
100
200
A_m / A_p
C_g = 30 mgr/m³
C_g = 10 mgr/m³
C_g = 3 mgr/m³

Fig. 3

CONCLUSION

In view of the basic importance of this effect the following suggestions are put forth:

Repetition of the HCHO-tests in the same C_g- and C_s-range

1.) with a much larger (5-10 x) particle size. [(ΔT) should vanish.]

2.) for one C_s-value at a different (ca. 10° - 15°) lower temperature. [(ΔT) should increase.]

3.) with (one) similar particle size of a substance (paraffin oil, (perhaps) polystyrene latex) which is chemically entirely inert toward the irritant used. (Similar, probably larger effects for (ΔT) would be expected and a closer similarity between HCHO and HCOOH.)

4.) with one C_s-value for a ca. 10 x larger "age" of the irritant-aerosol system. ((ΔT) should be affected to a degree depending on the affinity between irritant and particle substance.)

- - - - - - - -

August 1958